

**ENVIRONMENTAL MONITORING AT
ARGONNE NATIONAL LABORATORY**

ANNUAL REPORT FOR 1979

by

**N. W. Golchert, T. L. Duffy,
and J. Sedlet**



ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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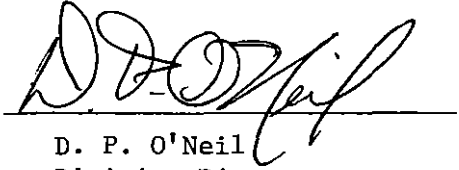
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ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1979 are presented and discussed. To evaluate the effect of Argonne operations on the environment, measurements were made for a variety of radionuclides in air, surface water, Argonne effluent water, soil, grass, bottom sediment, and foodstuffs; for a variety of chemical constituents in air, surface water, and Argonne effluent water; and of the environmental penetrating radiation dose. Sample collections and measurements were made at the site boundary and off the Argonne site for comparison purposes. Some on-site measurements were made to aid in the interpretation of the boundary and off-site data. The results of the program are interpreted in terms of the sources and origin of the radioactive and chemical substances (natural, fall-out, Argonne, and other) and are compared with applicable environmental quality standards. The potential radiation dose to off-site population groups is also estimated.

I. INTRODUCTION

A. General

This report is prepared to provide the U. S. Department of Energy (DOE) and the public with information on the levels of radioactive, chemical, and biological pollutants in the environment of Argonne National Laboratory (ANL) and on the amounts, if any, added to the environment as a result of Argonne operations. The report follows the guidelines given in DOE Manual Chapter 0513.⁽¹⁾ The Laboratory conducts a continuous environmental monitoring program on and near the Argonne site whose primary purpose is to determine the magnitude, origin, and identity of radioactive or potentially toxic chemical and biological substances in the environment. Of special interest is the detection of any such materials released to the environment by Argonne. One

important function of the program is to verify the adequacy of Argonne's pollution controls.

Argonne is a multi-disciplinary research and development laboratory with several principal objectives. It carries out a broad program of research activities in the physical, biomedical, and environmental sciences and serves as an important center for energy research and development, both nuclear and non-nuclear. Some of the energy-related research projects are nuclear reactor safety studies, improvements in the utilization of coal for power production, coal liquefaction studies, the development of electric batteries for vehicles and off-peak energy storage, magnetohydrodynamic power generation, solar energy utilization, and ocean thermal energy conversion using ammonia as the working fluid. Environmental research studies include a Great Lakes radioecology program, which is primarily concerned with the effects of effluents from nuclear and fossil fuel power plants on Lake Michigan and other watersheds, studies on the dispersion and behavior of airborne pollutants under various meteorological conditions, and reclamation of strip-mined lands. Almost all of the work at the Laboratory is of an unclassified nature.

The principal nuclear facilities at the Laboratory are a 5 MW heavy-water cooled and moderated general-purpose research reactor (CP-5) fueled with fully-enriched uranium, whose operation was terminated September 30, 1979; a 200 kW light-water cooled and moderated biological research reactor (Janus) fueled with fully-enriched uranium; one critical assembly or zero power reactor (ZPR-9), that is fueled at various times with plutonium, uranium, or a combination of the two; the Argonne Thermal Source Reactor (ATSR), a 10 kW research reactor fueled with enriched uranium; a 12.5 GeV proton accelerator, the Zero Gradient Synchrotron (ZGS), whose operation was also terminated September 30, 1979; a 60-inch cyclotron; several other charged particle accelerators (principally of the Van de Graaff type); ZING-P' (ZGS Intense Neutron Generator - Prototype); cobalt-60 irradiation sources; chemical and metallurgical plutonium laboratory; and several hot cells and laboratories designed for work with irradiated fuel elements and with multi-curie quantities of the actinide elements.

B. Description of Site

Argonne National Laboratory (Illinois site) occupies the central 6.88 sq.

km (1,700 acres) of a 15.14 sq km (3,740-acre) tract in DuPage County, 43 km (27 miles) southwest of downtown Chicago, and 39 km (24 miles) due west of Lake Michigan. It lies in the Des Plaines River Valley, south of Interstate Highway 55 and west of Illinois Highway 83. Figures 1 and 2 are maps of the site and of the surrounding area. The 8.26 km (2,040-acre) area surrounding the site (Waterfall Glen Forest Preserve) was formerly Argonne property, but was deeded to the DuPage County Forest Preserve District in 1973 for their use as a public recreational area, nature preserve, and demonstration forest.

The terrain is gently rolling, partially-wooded, former prairie and farmland. The grounds contain a number of small ponds and streams, the principal one being Sawmill Creek, which runs through the site in a southerly direction and enters the Des Plaines River about 2.1 km (1.3 miles) southeast of the center of the site. The land is drained primarily by Sawmill Creek, although the extreme southern portion drains directly into the Des Plaines River, which flows along the southern boundary of the Forest Preserve. This river flows southwest until it joins the Kankakee River about 48 km (30 miles) southwest of the Laboratory to form the Illinois River.

The largest topographical feature is the Des Plaines River channel, about 1.6 km (1 mile) wide. This channel contains the River, the Chicago Sanitary and Ship Canal, and the Illinois and Michigan Canal. Their presence extends the uninhabited area about 1.6 km (1 mile) south of the site. The elevation of the channel surface is 180 m (590 feet) above sea level. Bluffs, which comprise the south border of the site, rise from the channel at varying slope angles of 15° to 60°, reaching an average elevation of 200 m (650 feet) above sea level at the top. The land then slopes gradually upward reaching the average site elevation of 220 m (725 feet) above sea level at 940 m (3,000 feet) from the bluffs. Several large ravines oriented in a north-south direction are located in the southern portion of the site. The bluffs and ravines generally are forested with deciduous trees of an average height of 15-18 m (50-60 feet). The remaining portion of the site changes in elevation by no more than 7.6 m (25 feet) in a distance of 150 horizontal m (500 feet). In the southern portion of the Forest Preserve, the Chicago District Pipe Line Co. and the Atchison, Topeka, and Santa Fe Railroad have rights-of-way.

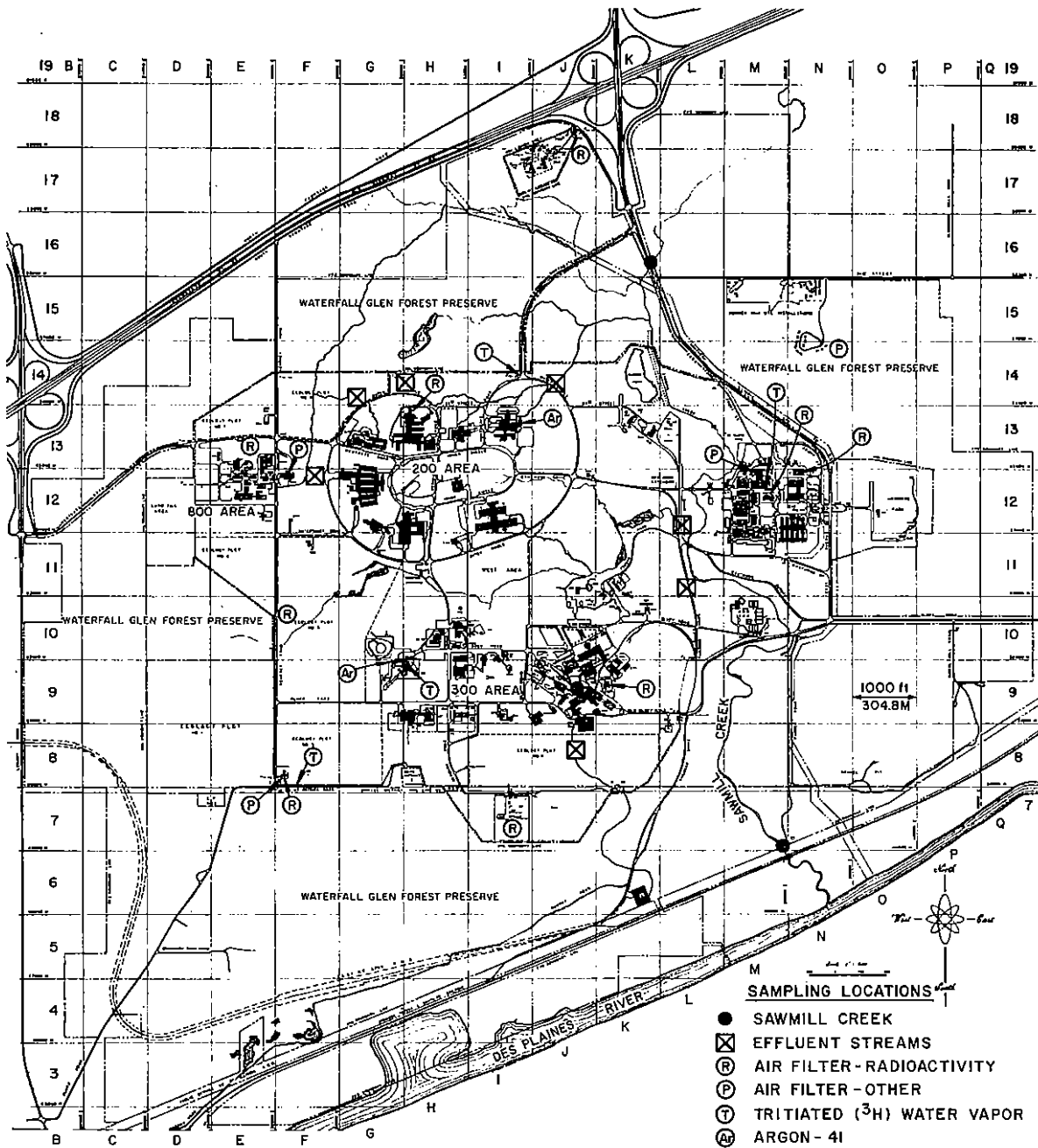


Fig. 1. Sampling Locations at Argonne National Laboratory

Fig. 2. Sampling Locations Near Argonne National Laboratory

C. Meteorology

The climate of the area is that of the upper Mississippi Valley, as moderated by Lake Michigan. A summary of the meteorological data collected on the site from 1950 to 1964 is available,⁽²⁾ and provides an adequate sample of the climate. Similar data have been collected since 1964.

The most important meteorological parameters for the purposes of this report are wind direction, wind speed, temperature, and precipitation. These are given here for 1979 and were provided by the Atmospheric Physics Section of the Radiological and Environmental Research Division at Argonne for the first half of the year and from the National Oceanic and Atmospheric Administration at Midway Airport for the second half of the year. The average monthly and annual wind roses are shown in Figure 3. The wind roses are polar coordinate plots in which the lengths of the radii represent the percentage frequency of wind speeds in classes of 2.01-6 m/s (4.5-13.4 mph), 6.01-10 m/s (13.4-22.4 mph), and greater than 10.01 m/s (22.4 mph). The direction of the radii represents the direction from which the wind blows. Sixteen radii are shown on each plot at 22.5° intervals; each radius represents the average wind speed for the direction covering 11.25° on either side of the radius. For example, in the plot labeled "79 total", the predominant wind blows from the south. The length and direction of this vector shows that, of the total wind observations, about 6% were in the 2.01-6 m/s range, about 3% were in the 6.01-10 m/s range, about 1% were greater than 10.01 m/s; and about 10% of the observations were from the direction between 168.75° and 191.25° (south). The number in the center represents the percent of observations of wind speed less than 2 m/s in all directions.

The roses show that the predominant winds were from the west, southwest, and south on the average, but seasonal variations are apparent. The winds were primarily westerly in January and February. In the spring, the northeast lake breezes are evident. The warm southerly winds, brought up by the high pressure areas in the southeastern U. S., began in May and continued for the remainder of the year. The winds during 1979 were characterized by a much stronger southerly component than the usual southwest dominance in past years. The winds are sufficiently variable so that monitoring for any airborne releases must be carried out in all directions from the site.

The precipitation and temperature data for 1979 are shown in Table 1.

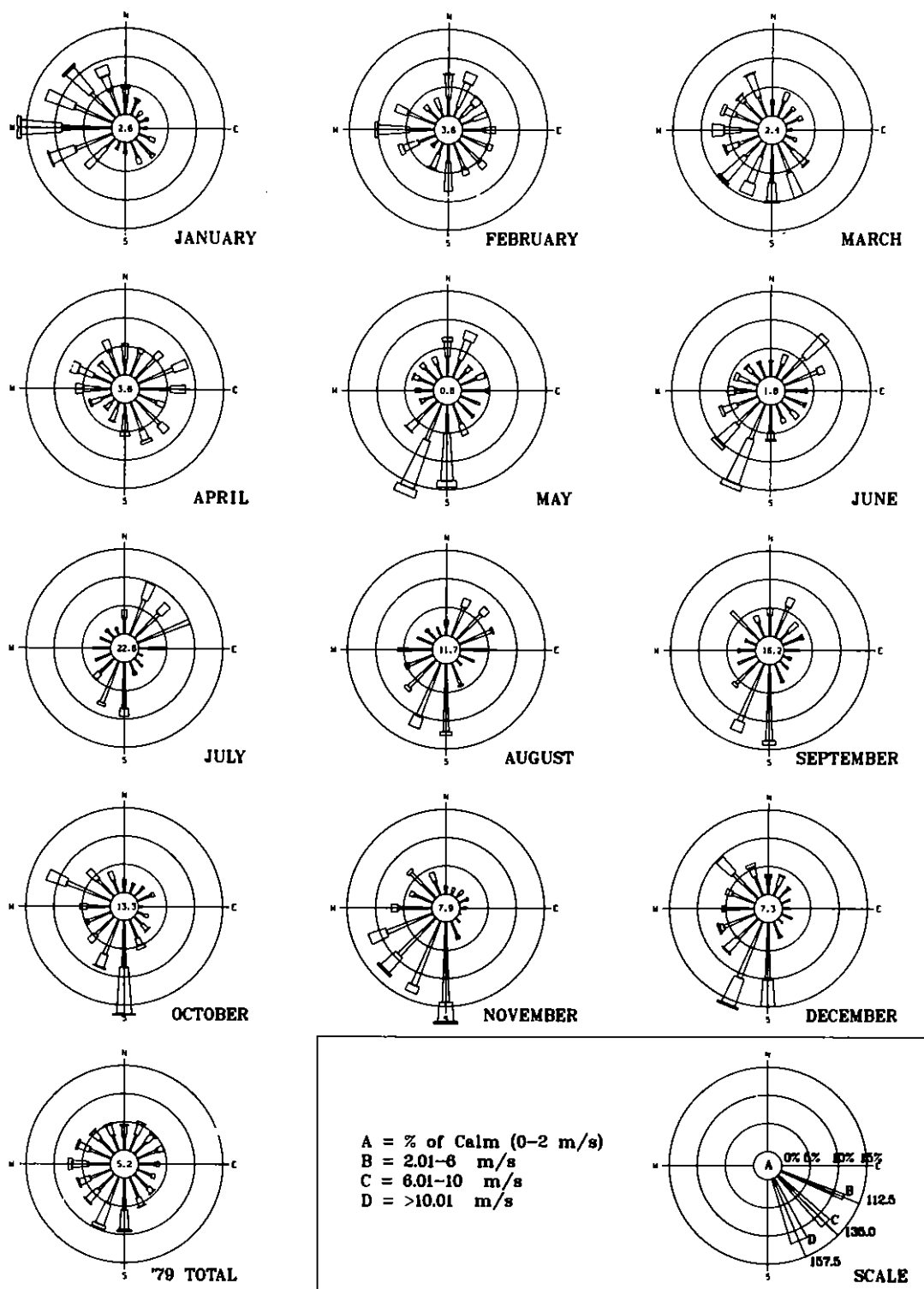


Fig. 3. Monthly and Annual Wind Roses at Argonne National Laboratory, 1979

Above-normal precipitation occurred in March, April, and August, while September was the driest month ever recorded for the area. Temperatures were below normal in January and February.

TABLE 1

Weather Summary, 1979

Month	Precipitation (cm)		Temperature (°C)	
	Amount	Average *	Monthly Average	Average **
January	7.52	4.88	-12.2	-4.2
February	3.25	4.57	-9.5	-2.7
March	12.55	6.93	2.0	2.5
April	11.91	7.98	7.2	8.8
May	6.15	8.81	15.0	14.6
June	6.58	9.47	20.2	20.2
July	8.18	8.66	21.5	23.2
August	21.9	8.10	22.0	22.5
September	0.03	8.08	18.8	18.7
October	4.60	6.55	12.1	12.5
November	7.52	5.88	4.7	4.7
December	7.30	5.30	1.2	-1.5

* Average precipitation, 1873-1977.

** Average temperature, 1871-1977.

D. Population

The area around Argonne has exhibited a large population growth in the past 15 years. Large areas of farmland have been converted into housing. A directional and annular 80-km (50-mile) population distribution for the area, which is used for the population dose calculations later in this report, is shown in Table 2.⁽³⁾ The distribution, centered on the CP-5 reactor, was obtained by modifying a similar distribution based on the 1970 U. S. Census and prepared by the Regional and Urban Studies Department at Oak Ridge National Laboratory. For distances within 8 km of the center, the updated values were largely based on quarter-section population data supplied by the Northeastern

TABLE 2

Incremental Population Data in the Vicinity of ANL, 1978

	In Thousands									
Distance, miles	0 - 1	1 - 2	2 - 3	3 - 4	4 - 5	5-10	10-20	20-30	30-40	40-50
Distance, km	0-1.6	1.6-3.2	3.2-4.8	4.8-6.4	6.4-8.0	8-16	16-32	32-48	48-64	64-80
<u>Direction</u>										
N	0	226	2171	3516	4859	44.0	163	309	168	193
NNE	0	124	1932	4314	2837	77.5	420	449	99	0
NE	0	347	1445	1233	1422	45.7	505	626	0	0
ENE	0	1729	3069	470	1617	45.0	684	289	0	0
E	0	9	212	0	12	19.2	653	508	13	26
ESE	0	0	88	275	118	18.6	213	320	308	46
SE	0	3	132	174	62	13.0	82	96	23	9
SSE	0	29	452	434	115	6.1	23	11	14	19
S	0	65	1305	688	772	3.9	31	4	27	40
SSW	0	39	3741	4557	665	11.8	97	8	18	7
SW	0	408	157	88	84	15.5	32	8	16	8
WSW	0	323	46	1199	2193	17.8	14	10	6	10
W	0	1242	740	7910	8852	13.4	42	18	15	8
WNW	0	662	136	2556	3960	24.5	73	46	6	52
NW	0	141	674	2267	6090	28.5	68	78	13	13
NNW	0	212	1390	1470	3414	37.0	101	135	91	66
Total	0	5559	17692	31151	37077	421.0	3202	2915	815	497
Cumulative Total	0		23251	54402	91479	512.0	3714	6629	7444	7941

Illinois Planning Commission (NIPC). Some adjustments were made on the basis of local observations. At distances beyond 8 km, but within the NIPC area, township forecasts were used, allocating population to each sector in proportion to the area of the township within the sector. The city of Chicago population was allocated in proportion to the area of each sector. Outside of the six-county area around Chicago included in the NIPC program, the Northwestern Indiana Regional Planning Commission provided quarter-section estimates for Lake and Porter Counties and other Illinois Counties provided estimates of the population in their respective areas. Population counts were usually based on locally adjusted estimates made by the State of Illinois Bureau of the Budget and on special interim U. S. censuses for several municipalities. Included within the 80-km radius are portions of Lake and Porter Counties, Indiana, portions of Kankakee, Grundy, LaSalle, DeKalb, McHenry, and Lake Counties in Illinois, and all of the DuPage, Will, Cook, Kendall, and Kane Counties in Illinois.

E. Water and Land Use

The principal stream that drains the site is Sawmill Creek. This Creek was formerly an intermittent stream, responding in flow rate largely to precipitation runoff. It now carries effluent water continuously from a municipal sewage treatment plant (Marion Brook Treatment Plant) located a few kilometers north of the site, which as an operation capacity of about 11.4 megaliters (3 million gallons) per day. In addition, the residential development in the area has resulted in the collection and channeling of additional runoff water into the Creek. Treated sanitary and laboratory waste water from Argonne are combined and discharged into Sawmill Creek at location 7M in Figure 1. This effluent averaged 2.7 megaliters (0.73 million gallons) per day, while the water flow in the Creek upstream of the waste-water outfall averaged about 41 megaliters (11 million gallons) per day during 1979. The combined Argonne effluent consisted of 65% laboratory waste water and 35% sanitary waste water.

During the middle part of 1979, the effluent channels from the 200 Area were combined and became part of the effluent sampled at 12L. The 12L effluent also drains the Freund ponds and enters Sawmill Creek at the same location. The stream from the 300 Area sampled at 11L is no longer in use. The stream sampled at 8J is still in use and this crosses the Laboratory boundary at 7K,

and soaks into the ground south of the site. Hence, the cooling tower effluents are now monitored at two locations, 8J and 12L, on a regular weekly basis, while the formerly used channels (11L, 12F, 14G, 14H, and 14J in Figure 1) are sampled monthly if they contain sufficient water. The latter measurement is to ascertain that unexpected chromate contamination does not occur.

Sawmill Creek and the Des Plaines River above Joliet, about 21 km (13 miles) southwest of Argonne, receive very little recreational or industrial use. A few people fish in these waters downstream from Argonne and some duck hunting takes place on the Des Plaines River. Water from the Chicago Sanitary and Ship Canal is used for some industrial purposes, such as hydroelectric generators and condensers, and for irrigation at the State prison near Joliet. It is also used as secondary cooling water by Argonne. The Canal, which carries Chicago Metropolitan Sanitary District effluent water, is used for industrial transportation and some recreational boating. Near Joliet, the River and Canal are combined into one waterway, which continues until it joins the Kankakee River to form the Illinois River about 48 km (30 miles) southwest of Argonne. The Dresden Nuclear Power Station complex is located at the confluence of the Kankakee, Des Plaines, and Illinois Rivers. This Station used water from the Kankakee for cooling, and discharges the water into the Illinois River. The first use of water for drinking is an indirect one at Alton, on the Mississippi River about 711 km (370 miles) downstream from Argonne, where water is used to replenish groundwater supplies by infiltration. In the vicinity of the Laboratory, only subsurface water (from all aquifers, shallow and deep) and Lake Michigan water are used for drinking purposes.

The principal recreational area near Argonne is Waterfall Glen Forest Preserve, which surrounds the site as described in Section I.B. and is shown in Figure 1. Most of this area has received little use thus far. It was available in 1979 for hiking, skiing, and equestrian sports, and its development and increased utilization by the public is expected. Very approximate estimates of usage are 600 individuals per day on weekends during the latter part of the year, after a new automobile parking lot was opened, and 500 individuals for group educational purposes during 1979. The average stay is about two hours. Sawmill Creek flows through the south portion of the Preserve on its way to the Des Plaines River. This region of the Preserve

(formerly named Rocky Glen) was used principally for picnicking, hiking, and overnight camping by youth groups, but has been closed for rehabilitation since June 6, 1976. East and southeast of Argonne and the Des Plaines River are located several large forest preserves of the Cook County Forest Preserve District. The preserves include the two sloughs shown in Figure 2, McGinnis and Saganashkee, as well as other smaller lakes. These areas are used for picnicking, boating, fishing, and hiking. A small park located in the eastern part of the Argonne site (12-0 in Figure 1) is for the use of Argonne and Department of Energy employees only.

The most recently available information on milk and principal agricultural production in a 10,600 sq km (4,100 sq mi) area around Argonne is shown by County in Table 3. These estimates were obtained from the Illinois Crop Reporting Service of the State Department of Agriculture and are for 1978.

TABLE 3

Agricultural Production Near ANL, 1978

County	Milk		Corn	Soybeans	Wheat	Oats
	No. of Cows	Million Pounds				
DuPage	200	2.1	2.1	0.64	0.10	0.073
Cook	200	2.1	1.6	0.74	0.098	0.13
Will	3,100	32.3	14.9	3.7	0.50	0.47
Kane	7,400	77	14.6	2.4	0.23	0.53
Kendall	600	6.2	9.8	1.9	0.15	0.18
Grundy	700	7.3	12.3	2.4	0.074	0.11
Lake	1,700	17.7	2.3	0.66	0.26	0.22

Note: To convert pounds into kilograms, multiply by 0.45; to convert bushels into cubic meters, multiply by 0.035.

II. SUMMARY

This is one in a series of annual reports prepared to provide the U. S. Department of Energy (DOE) and the public with information on the level of radioactive and chemical pollutants in the environment and on the amounts of such substances, if any, added to the environment as a result of Argonne operations. The previous report in this series is ANL-79-24. Included in this report are the results of measurements obtained in 1979 for a number of radionuclides in air, surface water, soil, grass, bottom sediment, and food-stuffs; for a variety of chemical constituents in air and water, and for the external penetrating radiation dose.

Total alpha and beta activities, fission and activation products, plutonium, thorium, and uranium were measured in air-filter samples collected continuously at the site perimeter and off the site. All the off-site and perimeter samples contained only radionuclides from natural sources and nuclear test detonations. Some short-lived fission products were detected at all sampling locations early in the year, and are attributed to fallout from the Chinese atmospheric nuclear test of December 14, 1978. No activity attributable to Argonne operations could be detected.

The plutonium-239,240 concentrations in air averaged 11×10^{-18} $\mu\text{Ci/ml}$,^{*} about a third of the 1978 value, and were nearly identical at all sampling locations. The monthly variations showed a "spring maximum" in stratospheric fallout of plutonium similar to that observed for beryllium-7 (a naturally-occurring nuclide) and several fission products. The results indicated that the airborne plutonium was from atmospheric nuclear test detonations and there was no evidence that any of the plutonium originated at Argonne. The average plutonium concentration was equivalent to 0.001% of the CG.^{**}

* The radioactivity units are described in Section III.

** Unless otherwise indicated, the hazard due to a given concentration of a radioactive nuclide is assessed in this report by comparison with the Concentration Guides (CG) and annual dose limits, or Radiation Protection Standards, for uncontrolled areas specified by the U. S. Department of Energy in Chapter 0524 of the DOE Manual.⁽⁴⁾ The pertinent CGs are listed in the Appendix, Section IV.B. Comparison with other standards is given where appropriate.

Argon-41 and hydrogen-3 (as water vapor) represent the major airborne radioactivity released from the Laboratory. The argon-41 concentration and corresponding radiation dose at the site boundary was less than the detection limit (50% of the CG) of the sampling system, but was calculated from an atmospheric dispersion model to be 1×10^{-10} $\mu\text{Ci/ml}$ and 1.2 mrem/yr in the predominant east to northeast directions to which the wind is blowing. These values are 0.2% of the CG and the non-occupational dose limit to individuals (500 mrem/yr) for uncontrolled areas. The calculated dose at 2.2 km (1.4 miles) NE, where the closest full-time residents live, was 0.6 mrem/yr. These values are consistent with penetrating radiation dose measurements made at the site perimeter. The measured hydrogen-3 concentration at the site perimeter averaged about 10×10^{-12} $\mu\text{Ci/ml}$, which is 0.005% of the CG and about eight times greater than the off-site concentration. The corresponding dose is 0.02 mrem/yr. These doses would only be received outdoors; no reduction for shielding by residences has been included.

Argonne waste water is discharged into Sawmill Creek, and this stream was sampled above and below the site to evaluate the effect of Argonne operations on its radioactive content. The nuclides (for which analyses were made) added to the Creek in the waste water, and the percent CG of their average Creek concentrations, were hydrogen-3, 0.012%; strontium-90, 0.11%; neptunium-237, 0.0002%; plutonium-239,240, 0.003%; americium-241, 0.0002%; and curium-244 and/or californium-249, 0.00002%. Although Sawmill Creek is not a source of potable water, the dose to an individual using water at these concentrations as his sole source of drinking water would be less than 0.5 mrem/yr.

Sawmill Creek flows into the Des Plaines River, which in turn flows into the Illinois River. The radioactivity levels in the latter two streams were similar to those in other streams in the area, and the activity added to the Creek by Argonne waste water had no measurable effect on the radioactive content of either the Des Plaines or Illinois Rivers.

Plutonium concentrations in soil showed the same general range and average at the site perimeter and off the site. The average plutonium-239,240 content of the top 5 cm of soil was 1.0×10^{-3} $\mu\text{Ci/m}^2$ at the site perimeter and 0.8×10^{-3} $\mu\text{Ci/m}^2$ off the site. The corresponding plutonium-238 averages were 0.07×10^{-3} $\mu\text{Ci/m}^2$ and 0.05×10^{-3} $\mu\text{Ci/m}^2$, respectively. The plutonium content of grass was similar to that found in previous years and was about a

factor of 10^4 less than soil from the same location. The results were within the range reported by other laboratories for fallout from test detonations and the plutonium found in soil and grass is attributed to this source. The plutonium content of samples from beds of streams and ponds ranged from 1×10^{-9} $\mu\text{Ci/g}$ to 8×10^{-9} $\mu\text{Ci/g}$ of plutonium-239,240, a range found in previous years to be normal for fallout plutonium in such materials. The concentrations of uranium, thorium, and several gamma-ray emitters measured in soil, plant, and bottom sediment were normal.

Milk from a dairy farm near the Laboratory was analyzed for several fission products, including hydrogen-3. Hydrogen-3 concentrations averaged $< 109 \times 10^{-9}$ $\mu\text{Ci/ml}$. The strontium-90 concentration (3.7×10^{-9} $\mu\text{Ci/ml}$) decreased by 15% compared to 1978, while the cesium-137 concentration (2.7×10^{-9} $\mu\text{Ci/ml}$) decreased by 25%.

Measurements of penetrating radiation were made at several locations at the site boundary and off the site. The off-site results averaged 89 mrem/yr with a standard deviation of 7 mrem/yr, which is in the normal range for the area. At three locations at the site boundary, above-normal readings were recorded that were attributable to Argonne operations. At two locations at the south fence the dose rates averaged, respectively, about 360 mrem/yr above normal (grid 7I in Figure 1) and 5-10 mrem/yr above normal (grid 8H) as a result of radiation from an on-site temporary storage facility for radioactive waste. About 300 m south of the fence, the measured dose rate dropped to 94 mrem/yr, which is within the normal range. Along the north side of the site, the dose at the fence at location 14I was about 55 mrem/yr above normal due to radiation from cobalt-60 sources in Building 202. Since all of these locations are unoccupied, there are no individuals receiving these measured doses. The calculated outdoor dose rate from these sources to the residents closest to the south boundary, about 1.6 km (1 mile) from the fence line, was about 0.04 mrem/yr; similarly the dose rate to the residents closest to the north boundary, about 0.75 km (0.5 mile) from the fence, was about 0.1 mrem/yr. Thus, doses to individuals near the site from these sources will not exceed 0.01% of the 500 mrem/yr limit.

Levels of chemical constituents and other water quality parameters were measured in Argonne waste and effluent water and in Sawmill Creek; the results were compared to the standards adopted by the State of Illinois. Concentra-

tions of mercury, hexavalent chromium, iron, and zinc in the Des Plaines River were measured to determine whether any contribution from Argonne waste water could be seen.

The weekly samples in the Argonne combined sanitary and laboratory waste-water effluent did not exceed the State of Illinois standard for Biochemical Oxygen Demand (B.O.D.) or suspended solids. Ammonia nitrogen exceeded the State standard in January, February, and December. Average concentrations of each of the other chemical constituents measured in this effluent were at or below these standards. The concentration of mercury averaged 78% of the State standard and exceeded this value 16% of the time (i.e., in 16% of the samples). No other constituent in this effluent exceeded the State standard at any time.

The concentrations of hexavalent chromium from six of the seven cooling tower effluent channels used for blowdown were below analytical detection limits. The levels in the effluent channel at 8J were consistently high. The effluent channels were combined in mid-1979 to yield two effluents, 8J and 12L. Zinc levels in the 12L effluent were consistently low.

The average values in Sawmill Creek for dissolved oxygen and chemical constituents, except ammonia nitrogen, dissolved solids, copper, and iron were within the State of Illinois standards. The average ammonia nitrogen level above the Argonne waste-water outfall was 1.5 times the State standard, and exceeded this standard in 59% of the samples. The average level below the outfall was 1.2 times the State standard and exceeded this value in 49% of the samples. The average concentration of dissolved solids above the Argonne waste-water outfall exceeded the State standard by 18%, but below the outfall it was less than the standard. The copper levels in 1979 below the outfall exceeded the State standard in 54% of the samples and averaged 1.8 times the standard. The levels in the Argonne effluent were insufficient to account for these high levels and studies indicated bottom sediment contamination to be a source. Individual values for iron, mercury, and silver exceeded State standards from 2% to 31% of the time. Hexavalent chromium levels were not in excess of the State standard at any time. The level of fecal coliform in the Argonne waste water did not exceed the individual sample standard of 400 organisms/100 ml, and at no time was the monthly standard of 200 organisms/100 ml (geometric mean) exceeded. Samples collected in the Des Plaines

River did not show any effect of Argonne effluent on levels of mercury, hexavalent chromium, iron, or zinc in the River.

Determination of total suspended particulates (TSP) in air was continued. Studies included iron, copper, lead, zinc, bromine, and strontium at the TSP locations for the first three months. Results for TSP were similar to last year. A 24-hour sample for TSP was established for the latter quarter of the year and values were somewhat lower than the comparable continuous sample.

III. MONITORING RESULTS

A. Radiological

The radioactivity of the environment was determined by measuring the concentrations of radioactive nuclides in naturally-occurring materials and by measuring the external penetrating radiation dose. Sample collections and measurements were made at the site perimeter and off the site principally for comparison purposes. Some on-site results are also reported when they are useful in interpreting perimeter and off-site results. Since radioactivity is usually spread by air and water, the sample collection program has concentrated on these media. In addition, soil, plants, foodstuffs, precipitation, and materials from the beds of lakes and streams were also collected and analyzed.

The results of radioactivity measurements are expressed in this report in terms of microcuries per milliliter ($\mu\text{Ci/ml}$) for water, air, and milk and microcuries per gram (g) and square meter (m^2) for soil and vegetation. When a nuclide was not detected, the result is given as less than ($<$) the minimum amount detectable (detection limit) by the analytical method used. Averages, including individual results that were less than the detection limit, were calculated by one of the following two methods. If a large fraction (usually 50% or more) of the individual results was less than the detection limit, the average was calculated with the assumption that such results were equal to the detection limit, and the resulting average value is expressed as less than ($<$) the computed average. If only a small fraction of the individual results was less than the detection limit, the average was calculated with the assumption that such results were actually one-half of the detection limit, and the average is given as a definite value. The former technique probably overestimates the average concentration in those samples below the detection limit and gives an upper limit for the average of all the samples in the group, since it is unlikely that all concentrations not detectable are at the detection limit. The latter method is based on the assumption that the values below the detection limit are distributed between zero and the detection limit with a frequency such that the average value is one-half of the detection limit. The averages that are obtained by using these two methods under the conditions indicated are believed to give an adequate

picture of the true average concentration at locations where the concentrations not only varied greatly, but were at times not detectable. Penetrating radiation measurements are reported in units of millirem (mrem) per year and population dose in man-rem.

Average values are usually accompanied by a plus-or-minus (\pm) limit value. Unless otherwise stated, this value is the 95% confidence limit calculated from the standard deviation of the average (standard error), and is a measure of the range in the concentrations encountered at that location. It does not represent the conventional error in the average of repeated measurements on the same or identical samples. Since many of the variations observed in environmental radioactivity are not random but occur for specific reasons (e.g., nuclear testing), samples collected from the same location at different times are not replicates. The more random the variation in activity at a particular location, the closer the confidence limits will represent the actual distribution of values at that location. The averages and confidence limits should be interpreted with this in mind. When a plus-or-minus figure accompanies an individual result in this report, it represents the statistical counting error at the 95% confidence level.

The measured concentration or radiation dose is compared with appropriate standards as a means of assessing the hazard. The standards used in this report are usually the Concentration Guides (CGs) and annual dose limits (Radiation Protection Standards) for uncontrolled areas given in DOE Manual Chapter 0524.⁽⁴⁾ The pertinent CGs as well as the detection limits are given in the Appendix, Section IV.B. Although the CGs apply to concentrations above natural levels, the percent of CG is sometimes given in this report for activities that are primarily of natural origin for comparative purposes. Such values are enclosed in parentheses to indicate this. Where other standards are used, their source is identified in the text.

1. Air

The radioactive content of particulate matter was determined by collecting and analyzing air-filter samples. The sampling locations are shown in Figures 1 and 2. Separate collections were made for specific radiochemical analyses and for alpha, beta, and gamma counting. The latter measurements were made on samples collected continuously on asbestos-cellulose filter

paper changed weekly at eight locations at the Argonne site perimeter* and at five locations off the site. Measurements were made at the perimeter because comparison between perimeter and off-site concentrations is necessary in evaluating and establishing the normal environmental concentration. If only off-site radioactivity were reported, their normality or origin could not be evaluated. Higher activities at the site perimeter may indicate radioactivity released by Argonne if the differences are greater than the error in sampling and measurement. Such results require investigation to determine the cause of the difference. The relative error is between 5 and 20% for most results, but approaches 100% at the detection limit.

The total alpha and beta activities in the individual weekly samples are summarized in Table 4. These measurements were made in low-background gas-flow proportional counters, and the counting efficiencies used to convert counting rates to disintegration rates were those measured for radon decay products on filter paper. The average concentrations of a number of gamma-ray emitters, as determined by gamma-ray spectrometry performed on composite weekly samples, are given in Table 5. The gamma-ray detector is a shielded 74 cm³ lithium-drifted germanium diode, calibrated for each gamma-ray emitting nuclide listed in Table 5.

The alpha activities, principally due to naturally-occurring nuclides, averaged the same as the past several years and were in their normal range. The average beta activity for the year, 34×10^{-15} $\mu\text{Ci/ml}$, was about a third of the 1978 average. The results in Table 5 indicate that the decrease was principally due to the decay of intermediate half-life fission products (zirconium-95-niobium-95, ruthenium-103, ruthenium-106-rhodium-106, cerium-141, and cerium-144) produced in previous nuclear test detonations. Residual fission products from the December 14, 1978, atmospheric nuclear test of the Peoples Republic of China, i.e., zirconium-95-niobium-95, ruthenium-103, barium-140-lanthanum-140, and cerium-141, were still evident early in 1979 in both the beta activity and gamma-ray emitters. No evidence of a "spring maximum" was apparent in the total beta activity whereas the more sensitive gamma-ray spectrometric measurements indicated this phenomenon. In addition to the cosmic-ray produced beryllium-7, the fission products ruthenium-106-rhodium-106, antimony-125, cesium-137, and cerium-144 exhibited increased air

*The site perimeter samplers are placed at the nearest location to the site boundary fence that provides electrical power and shelter.

TABLE 4

Total Alpha and Beta Activities in Air-Filter Samples, 1979*
(Concentrations in 10^{-15} $\mu\text{Ci/ml}$)

Month	Location	No. of Samples	Alpha Activity			Beta Activity		
			Avg.	Min.	Max.	Avg.	Min.	Max.
January	perimeter	36	2.0	0.7	3.6	49	35	76
	off-site	21	2.1	0.5	3.9	50	31	83
February	perimeter	34	2.3	0.8	3.3	46	17	72
	off-site	20	2.2	0.5	3.3	45	33	75
March	perimeter	38	1.3	0.5	2.0	33	22	44
	off-site	18	1.4	0.7	2.0	33	26	46
April	perimeter	38	1.6	0.5	2.6	33	18	50
	off-site	17	1.7	1.1	2.7	30	13	39
May	perimeter	40	1.9	1.0	3.2	33	20	43
	off-site	16	1.8	1.1	3.8	30	23	38
June	perimeter	42	2.0	0.9	3.9	35	21	45
	off-site	16	1.8	0.6	2.7	32	22	40
July	perimeter	39	1.8	0.5	4.2	33	16	48
	off-site	17	1.9	0.8	3.8	31	20	48
August	perimeter	39	1.6	0.7	3.0	26	14	40
	off-site	16	1.4	0.8	2.7	25	14	38
September	perimeter	39	2.3	0.9	4.5	31	17	52
	off-site	15	2.6	1.2	6.8	31	18	56
October	perimeter	39	1.9	0.6	5.0	24	15	35
	off-site	15	1.5	0.6	7.5	24	14	58
November	perimeter	39	2.0	0.8	3.8	34	19	64
	off-site	13	2.1	1.0	4.3	31	23	53
December	perimeter	39	1.8	0.8	3.9	28	17	38
	off-site	16	2.9	1.0	14	28	14	39
Annual Summary	perimeter	462	1.9 ± 0.2	0.5	5.0	34 ± 4	14	76
	off-site	200	2.0 ± 0.3	0.5	14	33 ± 4	13	83
Percent CG	perimeter	-	(0.002)	(0.0005)	(0.005)	0.034	0.014	0.076
	off-site	-	(0.002)	(0.0005)	(0.014)	0.033	0.013	0.083

* These results were obtained by measuring the samples four days after they were collected to avoid counting the natural activity due to short-lived radon and thoron decay products. This activity is normally present in the air and disappears within four days by radioactive decay.

TABLE 5

Gamma-Ray Activity in Air-Filter Samples, 1979

(Concentrations in 10^{-15} $\mu\text{Ci/ml}$)

Month	Location	^7Be	^{95}Zr - ^{95}Nb	^{103}Ru	^{106}Ru - ^{106}Rh	^{125}Sb	^{137}Cs	^{140}Ba - ^{140}La	^{141}Ce	^{144}Ce
January	perimeter	62	0.1	0.4	1.7	0.3	0.5	2.0	0.5	1.9
	off-site	70	0.2	0.4	1.8	0.4	0.7	1.6	0.5	2.1
February	perimeter	92	< 0.1	< 0.1	1.8	0.3	0.6	< 0.1	0.1	2.6
	off-site	95	< 0.1	< 0.1	2.0	0.4	0.7	< 0.1	< 0.1	2.6
March	perimeter	90	< 0.1	< 0.1	1.8	0.5	0.8	< 0.1	< 0.1	2.8
	off-site	88	< 0.1	< 0.1	2.2	0.4	0.8	< 0.1	< 0.1	3.2
April	perimeter	101	< 0.1	< 0.1	3.0	0.5	1.2	< 0.1	< 0.1	3.9
	off-site	93	< 0.1	< 0.1	2.3	0.5	1.1	< 0.1	< 0.1	3.6
May	perimeter	114	< 0.1	< 0.1	3.4	0.7	1.4	< 0.1	< 0.1	4.2
	off-site	105	< 0.1	< 0.1	2.8	0.7	1.4	< 0.1	< 0.1	4.1
June	perimeter	128	< 0.1	< 0.1	3.0	0.8	1.5	< 0.1	< 0.1	4.4
	off-site	112	< 0.1	< 0.1	2.7	0.7	1.3	< 0.1	< 0.1	3.9
July	perimeter	119	< 0.1	< 0.1	1.7	0.5	1.2	< 0.1	< 0.1	2.9
	off-site	111	< 0.1	< 0.1	2.0	0.4	1.2	< 0.1	< 0.1	3.1
August	perimeter	108	< 0.1	< 0.1	0.7	0.2	0.6	< 0.1	< 0.1	1.6
	off-site	98	< 0.1	< 0.1	1.1	0.4	0.5	< 0.1	< 0.1	1.6
September	perimeter	89	< 0.1	< 0.1	0.7	0.2	0.4	< 0.1	< 0.1	1.0
	off-site	88	< 0.1	< 0.1	0.9	0.3	0.4	< 0.1	< 0.1	1.0
October	perimeter	85	< 0.1	< 0.1	0.3	0.1	0.3	< 0.1	< 0.1	0.6
	off-site	82	< 0.1	< 0.1	0.6	0.2	0.3	< 0.1	< 0.1	0.8
November	perimeter	74	< 0.1	< 0.1	0.4	< 0.1	0.2	< 0.1	< 0.1	0.4
	off-site	74	< 0.1	< 0.1	0.5	0.2	0.2	< 0.1	< 0.1	0.4
December	perimeter	65	< 0.1	< 0.1	0.3	< 0.1	0.2	< 0.1	< 0.1	0.4
	off-site	74	< 0.1	< 0.1	0.2	< 0.1	0.3	< 0.1	< 0.1	0.4
Annual Summary	perimeter	94 ± 12	< 0.2	< 0.2	1.6 ± 0.6	0.4 ± 0.1	0.7 ± 0.3	< 0.3	< 0.2	2.2 ± 0.8
	off-site	91 ± 8	< 0.2	< 0.2	1.6 ± 0.5	0.4 ± 0.1	0.7 ± 0.2	< 0.3	< 0.2	2.2 ± 0.8
Percent CG ($\times 10^{-3}$)	perimeter	(0.24)	< 0.02	< 0.007	0.8	0.04	0.14	< 0.03	< 0.004	1.1
	off-site	(0.23)	< 0.02	< 0.007	0.8	0.04	0.14	< 0.03	< 0.004	1.1

concentrations in the spring. These nuclides were produced in past nuclear tests and their concentrations at the site perimeter and off the site were similar. About 95% of the gamma-ray activity was due to beryllium-7, produced in the stratosphere by cosmic-ray interactions.

The similarity of the annual averages of airborne alpha, beta, and gamma activities at the site perimeter and off the site indicates that these activities originated in a widespread source - fallout from nuclear test detonations and naturally-occurring materials - and not in a localized source such as Argonne.

Samples for radiochemical analyses were collected at perimeter locations 12N and 7I (Fig. 1) and off the site in Downers Grove (Fig. 2). Collections were made on a polystyrene filter medium. The total air volume filtered for the monthly samples was about 25,000 m³. Samples were ignited at 600°C to remove organic matter and prepared for analysis by vigorous treatment with hot hydrochloric, hydrofluoric, and nitric acids. This treatment has been found in our laboratory to solubilize plutonium that had been ignited at 1000°C.

Plutonium and thorium were separated on an anion exchange column. The acidity of the column effluent from this separation was adjusted to permit the extraction of uranium. Following the extraction, the aqueous phase was analyzed for radiostrontium by a standard radiochemical procedure. The separated plutonium, thorium, and uranium fractions were electrodeposited and measured by alpha spectrometry. The chemical recoveries were monitored by adding known amounts of plutonium-242, thorium-234, and uranium-232 tracers prior to ignition. Since alpha spectrometry cannot distinguish between plutonium-239 and plutonium-240, it should be understood that when plutonium-239 is mentioned in this report, the alpha activity due to the plutonium-240 isotope is also included. The results are given in Tables 6 and 7.

The average plutonium-239 concentrations decreased at all locations to about a third of the 1978 values. Figure 4 shows the monthly plutonium-239 air concentrations for the past seven years. The arrows at the bottom of the figure indicate the approximate dates of atmospheric nuclear tests. The average plutonium-238 concentration was similar to 1978, and the anticipated decrease was not apparent, probably due to the effect of the large measurement errors. The annual average strontium-90 concentrations decreased by about

TABLE 6

Strontium and Plutonium Concentrations in Air-Filter Samples, 1979
(Concentrations in 10^{-18} $\mu\text{Ci/ml}$)

Month	Location ¹	Strontium-89	Strontium-90	Plutonium-238	Plutonium-239 ²
January	12N	580 \pm 160	220 \pm 40	0.1 \pm 0.3	6.3 \pm 0.9
	7I	920 \pm 420	350 \pm 110	0.4 \pm 0.3	9.0 \pm 1.0
	off-site	330 \pm 180	170 \pm 60	0.3 \pm 0.3	5.8 \pm 0.8
February	12N	< 100	340 \pm 40	0.1 \pm 0.3	9.6 \pm 1.1
	7I	< 100	410 \pm 10	0.6 \pm 0.4	10.2 \pm 1.1
	off-site	< 100	280 \pm 30	0.6 \pm 0.5	7.5 \pm 1.1
March	12N	< 100	440 \pm 10	0.1 \pm 0.4	17 \pm 2
	7I	< 100	540 \pm 30	0.8 \pm 0.5	17 \pm 2
	off-site	< 100	380 \pm 100	1.0 \pm 1.1	14 \pm 2
April	12N	< 100	690 \pm 30	0.8 \pm 0.5	20 \pm 2
	7I	< 100	820 \pm 20	1.0 \pm 0.5	22 \pm 2
	off-site	< 100	590 \pm 30	0.3 \pm 0.3	15 \pm 1
May	12N	< 100	830 \pm 20	0.3 \pm 0.4	27 \pm 2
	7I	< 100	870 \pm 30	0.3 \pm 0.4	25 \pm 2
	off-site	< 100	730 \pm 20	0.3 \pm 0.8	27 \pm 3
June	12N	< 100	800 \pm 20	0.6 \pm 0.4	22 \pm 2
	7I	< 100	980 \pm 40	0.9 \pm 0.6	25 \pm 2
	off-site	< 100	840 \pm 20	0.7 \pm 0.4	22 \pm 2
July	12N	< 100	650 \pm 40	1.4 \pm 0.5	16 \pm 2
	7I	< 100	710 \pm 20	1.5 \pm 0.5	14 \pm 1
	off-site	< 100	520 \pm 10	1.4 \pm 0.5	12 \pm 1
August	12N	< 100	320 \pm 20	0.5 \pm 0.3	7.9 \pm 0.9
	7I	< 100	370 \pm 140	1.0 \pm 0.4	9.0 \pm 1.0
	off-site	< 100	-	0.5 \pm 0.3	6.4 \pm 0.8
September	12N	< 100	210 \pm 10	0.3 \pm 0.3	6.6 \pm 0.9
	7I	< 100	230 \pm 20	0.8 \pm 0.4	7.2 \pm 0.9
	off-site	< 100	190 \pm 30	0.5 \pm 0.4	5.0 \pm 0.9
October	12N	< 100	120 \pm 20	< 0.1	5.8 \pm 1.4
	7I	< 100	150 \pm 30	0.5 \pm 0.4	3.1 \pm 0.6
	off-site	< 100	100 \pm 20	0.6 \pm 0.4	2.2 \pm 0.6
November	12N	< 100	120 \pm 10	0.2 \pm 0.3	2.9 \pm 0.6
	7I	< 100	150 \pm 20	0.8 \pm 0.3	3.4 \pm 0.6
	off-site	< 100	80 \pm 40	0.4 \pm 0.5	4.6 \pm 0.7
December	12N	< 100	100 \pm 20	< 0.1	2.5 \pm 0.5
	7I	< 100	80 \pm 10	0.2 \pm 0.3	4.2 \pm 0.7
	off-site	< 100	90 \pm 20	0.3 \pm 0.3	1.6 \pm 0.4
Annual Summary	12N	< 140	400 \pm 160	0.4 \pm 0.2	12 \pm 5
	7I	< 170	470 \pm 180	0.7 \pm 0.2	12 \pm 5
	off-site	< 120	360 \pm 160	0.6 \pm 0.2	10 \pm 5
Percent CG ($\times 10^{-3}$)	12N	< 0.014	0.20	0.04	1.2
	7I	< 0.017	0.24	0.07	1.2
	off-site	< 0.012	0.18	0.06	1.0

¹Perimeter locations are given in terms of the grid coordinates in Figure 1

²Plutonium-240 is included (see text).

TABLE 7

Thorium and Uranium in Air-Filter Samples, 1979
(Concentrations in 10^{-18} $\mu\text{Ci/ml}$)

Month	Location ¹	Thorium-228	Thorium-230	Thorium-232 ²	Uranium-234	Uranium-235	Uranium-238 ²
January	12N	13 \pm 2	14 \pm 2	12 \pm 1	14 \pm 1	0.5 \pm 0.3	13 \pm 1
	7I	14 \pm 2	17 \pm 2	11 \pm 1	17 \pm 1	0.5 \pm 0.3	15 \pm 1
	off-site	15 \pm 2	17 \pm 2	13 \pm 1	11 \pm 1	0.2 \pm 0.2	11 \pm 1
February	12N	5 \pm 1	8 \pm 1	4 \pm 1	20 \pm 2	0.6 \pm 0.4	16 \pm 2
	7I	7 \pm 1	13 \pm 1	7 \pm 1	19 \pm 2	0.9 \pm 0.3	14 \pm 1
	off-site	7 \pm 2	3 \pm 1	1 \pm 1	7 \pm 1	0.2 \pm 0.4	9 \pm 2
March	12N	4 \pm 2	7 \pm 1	4 \pm 1	17 \pm 1	0.2 \pm 0.3	9 \pm 1
	7I	5 \pm 2	8 \pm 1	4 \pm 1	15 \pm 1	0.6 \pm 0.3	9 \pm 1
	off-site	3 \pm 2	5 \pm 1	3 \pm 1	6 \pm 1	0.5 \pm 0.4	6 \pm 1
April	12N	4 \pm 1	2 \pm 1	6 \pm 1	12 \pm 1	0.4 \pm 0.3	11 \pm 1
	7I	6 \pm 1	8 \pm 1	6 \pm 1	15 \pm 1	0.2 \pm 0.2	14 \pm 1
	off-site	5 \pm 1	6 \pm 1	4 \pm 1	6 \pm 1	0.3 \pm 0.2	7 \pm 1
May	12N	18 \pm 2	20 \pm 2	15 \pm 2	21 \pm 1	0.6 \pm 0.2	21 \pm 1
	7I	20 \pm 2	23 \pm 2	21 \pm 2	28 \pm 2	0.6 \pm 0.2	28 \pm 2
	off-site	12 \pm 2	13 \pm 2	11 \pm 1	18 \pm 2	0.5 \pm 0.3	18 \pm 2
June	12N	12 \pm 2	20 \pm 2	13 \pm 1	27 \pm 3	0.6 \pm 0.5	27 \pm 2
	7I	16 \pm 2	22 \pm 2	15 \pm 2	35 \pm 3	0.8 \pm 0.3	32 \pm 2
	off-site	20 \pm 2	24 \pm 1	14 \pm 1	24 \pm 2	1.0 \pm 0.3	25 \pm 2
July	12N	8 \pm 1	12 \pm 1	7 \pm 1	16 \pm 2	0.7 \pm 0.4	15 \pm 2
	7I	9 \pm 1	15 \pm 1	9 \pm 1	18 \pm 2	0.5 \pm 0.3	17 \pm 2
	off-site	11 \pm 1	16 \pm 1	11 \pm 1	17 \pm 1	0.4 \pm 0.3	11 \pm 1
August	12N	6 \pm 1	8 \pm 1	6 \pm 1	16 \pm 2	0.2 \pm 0.3	16 \pm 1
	7I	9 \pm 2	14 \pm 1	9 \pm 1	11 \pm 1	0.4 \pm 0.8	13 \pm 1
	off-site	4 \pm 1	5 \pm 1	3 \pm 1	8 \pm 1	0.2 \pm 0.6	7 \pm 1
September	12N	8 \pm 2	14 \pm 1	8 \pm 1	21 \pm 2	0.6 \pm 0.3	18 \pm 2
	7I	10 \pm 2	21 \pm 1	11 \pm 1	26 \pm 2	0.9 \pm 0.6	31 \pm 3
	off-site	3 \pm 2	4 \pm 1	3 \pm 1	15 \pm 2	1.0 \pm 0.5	12 \pm 2
October	12N	7 \pm 2	9 \pm 1	7 \pm 1	10 \pm 1	0.2 \pm 0.4	10 \pm 1
	7I	8 \pm 2	9 \pm 1	7 \pm 1	16 \pm 1	0.6 \pm 0.3	16 \pm 1
	off-site	4 \pm 2	4 \pm 1	4 \pm 1	7 \pm 1	0.5 \pm 0.3	6 \pm 1
November	12N	6 \pm 2	12 \pm 1	6 \pm 1	-	-	-
	7I	11 \pm 1	16 \pm 2	8 \pm 1	-	-	-
	off-site	4 \pm 2	5 \pm 1	3 \pm 1	-	-	-
December	12N	6 \pm 2	12 \pm 1	6 \pm 1	21 \pm 2	0.5 \pm 0.3	19 \pm 2
	7I	13 \pm 3	16 \pm 2	15 \pm 2	21 \pm 2	0.8 \pm 0.3	19 \pm 1
	off-site	7 \pm 2	9 \pm 1	7 \pm 1	10 \pm 1	0.4 \pm 0.3	10 \pm 1
Annual Summary	12N	8 \pm 2	12 \pm 3	8 \pm 2	18 \pm 3	0.5 \pm 0.1	16 \pm 3
	7I	11 \pm 3	15 \pm 3	10 \pm 3	20 \pm 4	0.6 \pm 0.1	19 \pm 5
	off-site	8 \pm 3	9 \pm 4	6 \pm 3	12 \pm 4	0.5 \pm 0.2	11 \pm 3
Percent CG ($\times 10^{-3}$)	12N	(4.0)	(4.0)	(0.8)	(0.45)	(0.012)	(0.32)
	7I	(5.5)	(5.0)	(1.0)	(0.50)	(0.015)	(0.38)
	off-site	(4.0)	(3.0)	(0.6)	(0.30)	(0.012)	(0.22)

¹Perimeter locations are given in terms of the grid coordinates in Figure 1.

²The concentrations in units of $\mu\text{g/m}^3$ can be obtained by multiplying the value in $\mu\text{Ci/ml}$ by 2.96×10^{12} for uranium-238 and by 9×10^{12} for thorium-232. The mass of the other thorium isotopes in comparison to thorium-232 and the other uranium isotopes in comparison to uranium-238 is negligible.

PLUTONIUM-239, 240 AIR CONCENTRATION

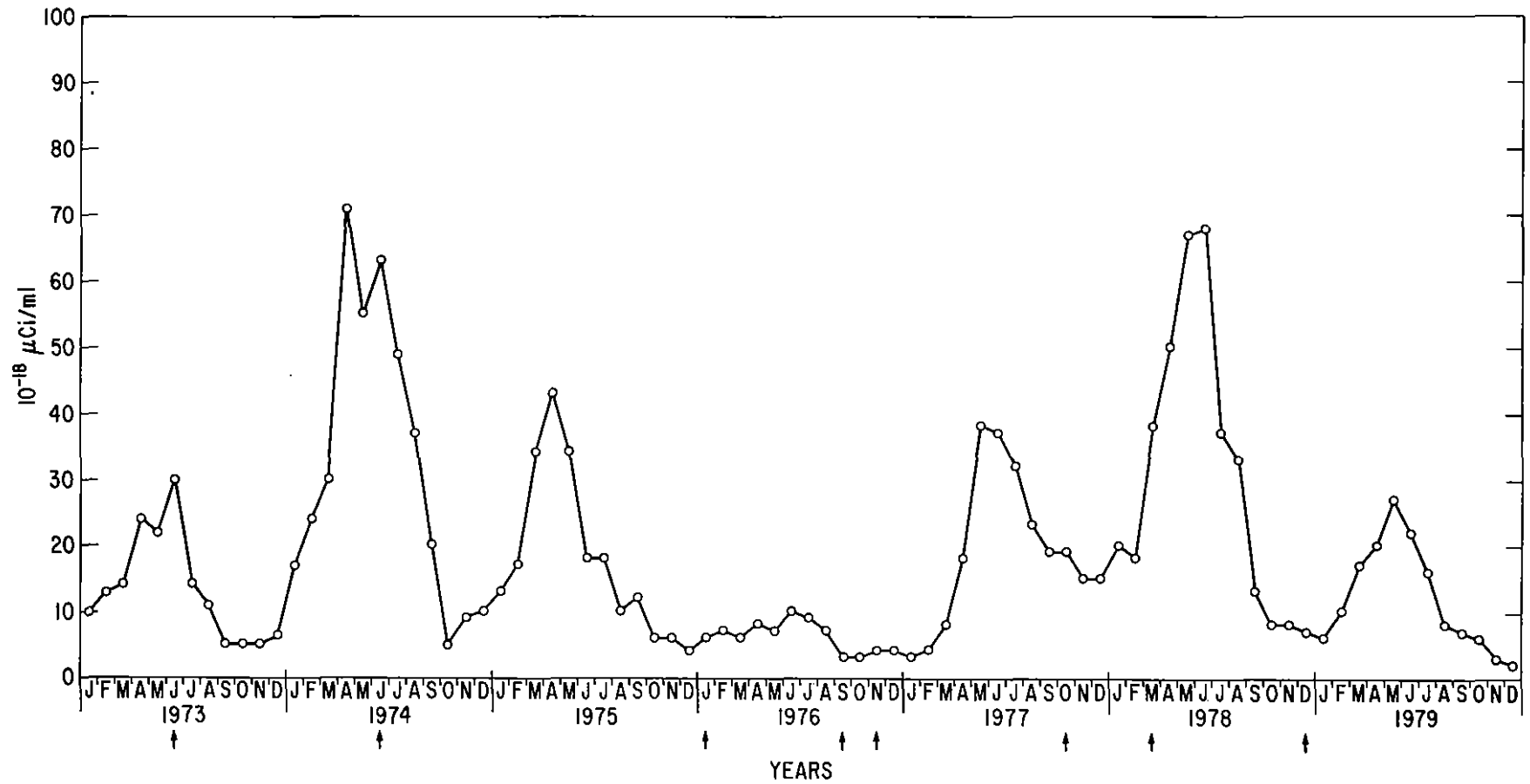


Fig. 4. Plutonium-239, 240 Air Concentrations, 1973-79

the same rate as the plutonium-239, while the strontium-89 was only detectable during January, as discussed previously. The long-lived radionuclides produced in nuclear detonations, plutonium-239, strontium-90, and cesium-137, showed similar monthly variations. There is no indication that these radionuclides originated from Argonne operations. Their concentrations were similar at the site perimeter and off the site and a spring peak is evident, indicating their stratospheric-fallout origin.

The thorium and uranium concentrations are in the same range found during the past several years and are considered to be of natural origin. The percent of CG for the averages is included for completeness; the values are placed in parentheses since the concentrations are considered to be background levels. The amounts of thorium and uranium in a sample were proportional to the mass of material collected on the filter paper, and the bulk of these elements in the air was due to resuspension of soil. In contrast, the amount of plutonium in the air samples contributed by soil, if the resuspended soil has the same plutonium concentration as the first centimeter on the ground, ranged from about 1.2% in March to 13% in November of the total plutonium in the samples.

Air sampling for argon-41 and hydrogen-3 (tritiated) water vapor was conducted in the exhaust stack of the CP-5 reactor (Building 330, 9H) because this reactor was the principal source of these nuclides at the Laboratory, and measurement of their concentrations at this location provides a source term. On September 30, 1979, operation of the CP-5 reactor was terminated. As a result, no argon-41 was generated from this source during the last quarter of the year, but tritiated water vapor continued to be released because the heavy water moderator was not removed until the end of the year. Argon-41 was collected by filling an evacuated "Marinelli-type" container with air and was measured by gamma-ray spectrometry. Hydrogen-3 (tritiated) water vapor in air was collected by adsorption on silica gel and was measured by counting the desorbed water in a liquid scintillation counter.

Hydrogen-3 concentrations at three perimeter locations and off-site are given in Table 8. The 12M location (1900 m east-northeast of the CP-5 reactor) may be considered a perimeter sample for CP-5. The average hydrogen-3 concentrations at all perimeter locations are about 40% lower than in 1978. The data show correlation with wind direction and indicate that, relative to

TABLE 8

Hydrogen-3 Perimeter and Off-Site Concentrations, 1979

Month	Location *	No. of Samples	Concentrations in 10^{-12} $\mu\text{Ci/ml}$		
			Avg.	Min.	Max.
January	8F	9	1.3	0.1	7.0
	14I	9	0.8	0.1	1.7
	12M	2	2.0	1.5	2.5
	off-site	2	0.4	0.3	0.5
February	8F	8	4.2	0.3	8.2
	14I	8	2.3	0.6	7.8
	12M	2	2.3	1.2	3.4
	off-site	2	1.8	0.4	3.2
March	8F	9	13	0.1	67
	14I	9	3.7	1.6	8.4
	12M	2	4.8	3.5	6.2
	off-site	2	0.5	0.4	0.7
April	8F	8	11	0.1	35
	14I	8	3.1	0.4	11
	12M	2	2.8	0.6	5.1
	off-site	2	0.6	0.6	0.7
May	8F	9	12	1.6	70
	14I	9	12	2.1	31
	12M	2	7.4	1.0	14
	off-site	2	1.0	0.8	1.1
June	8F	9	38	3.7	109
	14I	9	11	2.8	22
	12M	2	3.8	1.8	5.8
	off-site	2	1.1	0.3	1.8
July	8F	9	26	3.2	82
	14I	9	10	2.7	30
	12M	2	15	9.4	21
	off-site	2	2.1	1.9	2.2
August	8F	8	30	2.4	122
	14I	8	15	2.0	42
	12M	2	4.6	3.2	6.0
	off-site	2	1.5	1.4	1.7
September	8F	9	22	2.5	92
	14I	9	34	2.5	142
	12M	2	8.6	6.7	11
	off-site	2	2.6	2.4	2.7
October	8F	9	5.3	0.1	39
	14I	9	7.3	0.8	20
	12M	2	8.6	0.6	17
	off-site	2	0.5	0.3	0.7
November	8F	8	4.6	0.5	14
	14I	8	4.4	1.6	10
	12M	2	4.3	0.7	7.8
	off-site	2	1.7	1.1	2.3
December	8F	8	2.0	0.3	6.8
	14I	8	9.3	0.1	31
	12M	2	7.1	0.6	14
	off-site	2	0.4	0.4	0.4
Annual Summary	8F	103	14 \pm 7	0.1	122
	14I	103	9.3 \pm 5.1	0.1	142
	12M	24	5.9 \pm 2.1	0.6	21
	off-site	24	1.2 \pm 0.4	0.3	3.2
Percent CG	8F	-	0.0070	0.00005	0.061
	14I	-	0.0045	0.00005	0.071
	12M	-	0.0030	0.00030	0.010
	off-site	-	0.0006	0.00015	0.0016

* Perimeter locations are given in terms of the grid coordinates in Figure 1.

the reactor, dilution to the background level occurs before reaching the site boundary in directions other than that from which the wind is blowing. The average and maximum perimeter concentrations were equivalent to 0.005% and 0.07% of the CG, respectively.

The off-site concentrations, measured about 10 km (6.2 miles) northwest of the Laboratory, were also 40% lower than levels observed at this location in 1978. This background level of hydrogen-3 should be subtracted from the other concentrations in Table 8 to obtain the Argonne contribution.

Argon-41 and hydrogen-3 (in the form of tritiated water) from the CP-5 reactor constitute the major portion of the gaseous radioactive effluent released from the Laboratory. During the first nine months of 1979 when CP-5 was operating, the total amount of argon-41 discharged from the reactor was estimated to be 7.1×10^3 Ci, based on a measured release rate of 1.03 Ci/MW-hr. Since the half-life of this nuclide is only 110 minutes, about 5% will decay before reaching the site boundary if the argon-41 moves with an average wind speed of 3.4 m/s (7.6 mph). The total amount of hydrogen-3 (as tritiated water) discharged from the CP-5 reactor was 660 Ci. These discharges and the corresponding doses will be discussed further in Section III.A.6.a.

Other airborne effluents were considerably lower. A small amount of argon-41, about 0.8 Ci in 1979, was released from the Janus reactor (Building 202, location 13I). The other major effluent was krypton-85, estimated to be 9 Ci. Several other fission products were also released in millicurie or smaller amounts. The release of iodine-131 (a nuclide of particular interest) in CP-5 exhaust air was estimated to be 0.012 Ci, based on concentration measurements in the stack. The maximum concentration at 1.5 km (0.93 mile), assuming no ground deposition, would be about 1×10^{-16} μ Ci/ml, or $10^{-4}\%$ of the CG.

2. Surface Water

Total (nonvolatile) alpha and beta activities were determined by counting the residue remaining after evaporation of the water, and applying counting efficiency corrections determined for uranium-233 (for alpha activity) and thallium-204 (for beta activity) to obtain disintegration rates. Hydrogen-3 was determined by liquid scintillation counting of a separate sample, and this activity does not appear in the total nonvolatile beta activity. Uranium

was determined fluorophotometrically, and the results calculated in terms of activity with the assumption that the isotopic composition was that of natural uranium. Analyses for other radionuclides were performed by specific radiochemical separations followed by appropriate counting. One liter aliquots were used for all analyses except hydrogen-3 and the transuranium nuclides. Hydrogen-3 analyses were performed by counting 10 ml in a gel system. Analyses for transuranium nuclides were performed on 10 or 50-liter samples by chemical separation methods followed by alpha spectrometry.^(5,6) Plutonium-236 was used to determine the yields of plutonium and neptunium, which were separated together. A group separation of a fraction containing the transplutonium elements was monitored for recovery with americium-243 tracer.

Argonne waste water is discharged into Sawmill Creek, a small stream that runs through the Laboratory grounds, drains surface water from much of the site, and flows into the Des Plaines River about 500 m (0.3 mile) downstream from the waste-water outfall. Sawmill Creek was sampled upstream from the Argonne site and downstream from the waste-water outfall to determine if radioactivity was added to the stream by Argonne waste water or from surface drainage. The sampling locations are shown in Figure 1. Below the waste-water outfall, daily samples were collected by a continuous sampler, which operated about 77% of the year. When the continuous sampling device was not functioning, a grab sample was collected each working day. Equal portions of the daily samples collected each week were combined and analyzed to obtain an average weekly concentration. Above the site, samples were usually collected once a month and were analyzed for the same radionuclides as the below-outfall samples.

Annual summaries of the results obtained for Sawmill Creek are given in Table 9. Comparison of the results, and 95% confidence limits of the averages, for the two sampling locations shows that the nuclides whose presence in Creek water can be attributed to Argonne operations were hydrogen-3, neptunium-237, plutonium-239, americium-241, and occasionally strontium-90, plutonium-238, curium-242 and/or californium-252, and curium-244 and/or californium-249. The percentage of individual samples containing activity attributable to Argonne was 100% for plutonium-239; 98% for neptunium-237; 70% for americium-241; and 90% for hydrogen-3. The concentrations of all these nuclides were low compared to the CGs. The principal radionuclide added to the Creek by

TABLE 9

Radionuclides in Sawmill Creek, 1979

Type of Activity	Location *	No. of Samples	Concentration (10^{-9} $\mu\text{Ci/ml}$)			Percent CG		
			Avg.	Min.	Max.	Avg.	Min.	Max.
Alpha (nonvolatile)	16K	12	2.3 ± 0.7	0.8	4.4	(0.077)	(0.027)	(0.15)
	7M	251	1.6 ± 0.3	0.4	5.7	(0.053)	(0.013)	(0.19)
Beta (nonvolatile)	16K	12	20 ± 4	10	32	(0.67)	(0.33)	(1.07)
	7M	251	17 ± 2	7	30	(0.57)	(0.23)	(1.00)
Hydrogen-3	16K	12	120 ± 42	< 100	263	0.0040	< 0.003	0.0088
	7M	251	362 ± 60	< 100	1000	0.012	< 0.003	0.033
Strontium-89	16K	12	-	-	< 2	-	-	< 0.07
	7M	251	-	-	< 2	-	-	< 0.07
Strontium-90	16K	12	< 0.28	< 0.25	0.50	< 0.093	< 0.08	0.17
	7M	251	0.32 ± 0.17	< 0.25	0.72	0.11	< 0.08	0.24
Iodine-131	16K	12	< 4.7	< 3	23	< 1.6	< 1	7.7
	7M	251	< 3.1	< 3	3.8	< 1.0	< 1	1.3
Barium-140	16K	12	-	-	< 2	-	-	< 0.007
	7M	122	-	-	< 2	-	-	< 0.007
Uranium **	16K	12	2.0 ± 0.3	1.5	2.8	(0.0050)	(0.0038)	(0.0070)
	7M	251	1.7 ± 0.4	1.0	2.9	(0.0042)	(0.0025)	(0.0072)
Neptunium-237	16K	12	-	-	< 0.001	-	-	< 0.00003
	7M	251	0.0055 ± 0.0020	< 0.001	0.046	0.00018	< 0.00003	0.0015
Plutonium-238	16K	12	-	-	< 0.001	-	-	< 0.00002
	7M	251	< 0.0015	< 0.001	0.0047	< 0.00003	< 0.00002	0.00094
Plutonium-239	16K	12	< 0.00052	< 0.0005	0.00068	< 0.000010	< 0.00001	0.000014
	7M	251	0.015 ± 0.004	0.0012	0.059	0.00030	0.000024	0.0012
Americium-241	16K	12	< 0.0011	< 0.001	0.0012	< 0.000028	< 0.000025	0.000030
	7M	251	0.0098 ± 0.0038	< 0.001	0.065	0.00024	< 0.000025	0.0016
Curium-242 and/or Californium-252	16K	12	-	-	< 0.001	-	-	< 0.000005
	7M	251	< 0.0014	< 0.001	0.0092	< 0.000007	< 0.000005	0.000046
Curium-244 and/or Californium-249	16K	12	-	-	< 0.001	-	-	< 0.000014
	7M	251	< 0.0048	< 0.001	0.044	< 0.000069	< 0.000014	0.00063

* Location 16K is upstream from the Argonne site and location 7M is downstream from the Argonne waste-water outfall.

** Uranium concentrations in units of $\mu\text{g/l}$ can be obtained by multiplying the concentration given by 1.48×10^9 .
The average concentration in the Creek then becomes $3.0 \mu\text{g/l}$.

Argonne waste water, in terms of concentration, was hydrogen-3. Its average net concentration (equivalent to 0.01% of the CG) was the lowest since such measurements were made. However, the total concentration, regardless of source, must be used in assessing the hazard of a radionuclide not naturally present, so the percent CG in the table was calculated on this basis.

The hydrogen-3 in the Creek above the site was similar in concentration to levels found away from the Laboratory site and is characteristic of the current ambient levels in surface water. During 1979, the hydrogen-3 content of other lakes and streams ranged from $< 100 \times 10^{-9}$ $\mu\text{Ci/ml}$ to 360×10^{-9} $\mu\text{Ci/ml}$ and averaged 130×10^{-9} $\mu\text{Ci/ml}$.

The average total alpha and beta activities were slightly higher above the site, which indicates that at times Argonne waste water contained less of these materials than Creek water. The higher activities above the site were probably due to the water added to the Creek by a large municipal sewage treatment plant. The large amount of dissolved solids present in the sewage water is naturally accompanied by a small amount of radioactive materials, and increases the radioactivity in the Creek water.

In addition to the natural beta activity and that added by Argonne waste water at the outfall, beta activity from nuclear detonations was detected at both sampling locations. The normal nonvolatile beta activity is approximately 8×10^{-9} $\mu\text{Ci/ml}$, while the contribution from the upstream municipal sewage treatment plant is another 8×10^{-9} $\mu\text{Ci/ml}$. It is estimated that fallout activity added about 1×10^{-9} $\mu\text{Ci/ml}$ to the nonvolatile beta activity at both locations and that the Argonne contribution to the water below the outfall averaged about 1×10^{-9} $\mu\text{Ci/ml}$. The Argonne contribution remained the same as in 1978, while the fallout contribution decreased by a factor of two.

The total radioactive effluent discharged to the Creek in Argonne waste water can be estimated from the average concentrations and the volume of water carried by the Creek. These totals are 3.8 Ci of hydrogen-3, 0.6 mCi of strontium-90, 0.2 mCi of plutonium-239, 0.1 mCi of americium-241, 0.07 mCi of neptunium-237, and < 0.05 mCi of curium and californium nuclides.

Since Sawmill Creek empties into the Des Plaines River, which in turn flows into the Illinois River, the radioactivity in the latter two streams is

important in assessing the contribution of Argonne waste water to the environmental radioactivity. The Des Plaines River was sampled twice a month below, and monthly above, the mouth of Sawmill Creek to determine if the radioactivity in the Creek had any effect on the activity in the River. Annual summaries of the results obtained for these two locations are given in Table 10. The average nonvolatile alpha, beta, and uranium concentrations in the River were very similar to past averages and remain in the normal range. Results were quite similar above and below the Creek for all radionuclides since the activity in Sawmill Creek was reduced by dilution so that it was not detectable as such in the Des Plaines River. The natural nonvolatile beta activity was about 12×10^{-9} $\mu\text{Ci/ml}$, and the excess, 1×10^{-9} $\mu\text{Ci/ml}$, was due to fallout. The average nonvolatile alpha and beta activities, 1.6×10^{-9} $\mu\text{Ci/ml}$ and 11×10^{-9} $\mu\text{Ci/ml}$, respectively, of 21 off-site surface water samples collected this year (excluding the Des Plaines River) were similar to the levels found in previous years.

On occasion, detectable concentrations of iodine-131 were found at the Sawmill Creek and/or Des Plaines River sampling locations. No iodine-131 was found at any other location or in air-filter samples. The source may be a medical facility or a sewage treatment plant that has processed an excretion from an individual who has received a medical injection of iodine-131. Although the presence of iodine-131 is infrequent and unpredictable, an attempt will be made to identify the source or sources of the iodine-131.

The radioactivity in samples of Illinois River water, shown in Table 11, was similar to those found in other bodies of water in the area and to the activities found previously at these same locations. No radioactivity originating at Argonne could be detected in the Des Plaines or Illinois Rivers.

3. Soil, Grass, and Bottom Sediment

The plutonium content of soil, grass, and bottom sediment was measured at the site perimeter and off the site. The object of the off-site sampling was to determine the deposition of plutonium from weapons testing for comparison with perimeter samples, and with results obtained by other organizations for samples collected at large distances from nuclear installations. This latter comparison is useful in determining if the soil activity near Argonne is normal. For this purpose, the site selection criteria and sample

TABLE 10

Radionuclides in Des Plaines River Water, 1979

Type of Activity	Location *	No. of Samples	Concentration (10^{-9} $\mu\text{Ci/ml}$)			Avg.	Percent CG	
			Avg.	Min.	Max.		Min.	Max.
Alpha (nonvolatile)	A	12	1.4 ± 0.3	0.6	2.3	(0.047)	(0.020)	(0.077)
	B	23	1.6 ± 0.1	1.0	2.2	(0.053)	(0.033)	(0.073)
Beta (nonvolatile)	A	12	11 ± 2	5.5	18	(0.37)	(0.18)	(0.60)
	B	23	13 ± 2	5.6	22	(0.43)	(0.19)	(0.73)
Hydrogen-3	A	12	< 133	< 100	186	< 0.004	< 0.003	0.0062
	B	23	< 126	< 100	206	< 0.004	< 0.003	0.0069
Strontium-89	A	12	-	-	< 2	-	-	< 0.07
	B	23	-	-	< 2	-	-	< 0.07
Strontium-90	A	12	0.39 ± 0.06	0.25	0.54	0.13	0.083	0.18
	B	23	0.40 ± 0.04	< 0.25	0.54	0.13	< 0.08	0.18
Iodine-131	A	12	< 5.1	< 3	26	< 1.7	< 1	8.7
	B	23	< 3.7	< 3	15	< 1.2	< 1	5.0
Barium-140	A	12	-	-	< 2	-	-	< 0.007
	B	11	-	-	< 2	-	-	< 0.007
Uranium ** (natural)	A	12	1.6 ± 0.4	0.7	2.7	(0.0040)	(0.0018)	(0.0068)
	B	23	1.6 ± 0.3	0.6	3.0	(0.0040)	(0.0015)	(0.0075)
Neptunium-237	A	11	< 0.0011	< 0.001	0.0015	< 0.000036	< 0.00003	0.000050
	B	12	< 0.0011	< 0.001	0.0011	< 0.000036	< 0.00003	0.000036
Plutonium-238	A	11	-	-	< 0.001	-	-	< 0.00002
	B	12	-	-	< 0.001	-	-	< 0.00002
Plutonium-239	A	11	< 0.00056	< 0.0005	0.00079	< 0.000011	< 0.00001	0.000016
	B	12	< 0.00072	< 0.0005	0.0022	< 0.000016	< 0.00001	0.000044
Americium-241	A	12	-	-	< 0.001	-	-	< 0.000025
	B	12	-	-	< 0.001	-	-	< 0.000025
Curium-242 and/or Californium-252	A	12	-	-	< 0.001	-	-	< 0.000005
	B	12	-	-	< 0.001	-	-	< 0.000005
Curium-244 and/or Californium-249	A	12	-	-	< 0.001	-	-	< 0.000014
	B	12	-	-	< 0.001	-	-	< 0.000014

* Location A, near Route 45, is upstream and location B, near Lemont, is downstream from the mouth of Sawmill Creek. See Figure 2.

** Uranium concentrations in units of $\mu\text{g/l}$ can be obtained by multiplying the concentration given by 1.48×10^9 . The average concentration is $2.4 \mu\text{g/l}$.

TABLE 11

Radionuclides in Illinois River Water, 1979
(Concentrations in 10^{-9} $\mu\text{Ci/ml}$)

Location	Date Collected	Alpha *	Beta *	Hydrogen-3	Uranium ** (natural)	Neptunium-237	Plutonium-239
McKinley Woods State Park	June 5	1.5	14	107	1.4	< 0.001	0.0022
Below Dresden Power Station	June 5	0.8	7.0	< 100	1.3	< 0.001	0.0019
Morris	June 5	1.2	12	< 100	1.4	-	-
Starved Rock State Park	June 5	1.2	7.6	< 100	1.5	-	-
McKinley Woods State Park	October 9	0.8	8.6	161	0.5	< 0.001	< 0.0001
Below Dresden Power Station	October 9	0.6	8.1	240	0.6	< 0.001	0.0001
Morris	October 9	1.0	7.2	184	0.8	-	-
Starved Rock State Park	October 9	2.2	8.6	160	1.0	-	-

*Nonvolatile activity.

** Uranium concentrations in units of $\mu\text{g/l}$ can be obtained by multiplying the concentration by 1.48×10^9 .

preparation techniques of the DOE Environmental Measurements Laboratory⁽⁷⁾ were used. Sites were selected in several directions and at various distances from the Laboratory. Each site was selected on the basis that the soil appeared, or was known to have been, undisturbed for a number of years. Attempts were made to select open, level, grassy areas that were mowed at reasonable intervals. Public parks were selected when available.

Each soil sample consisted of five cores totaling 432 cm² in area by 5 cm deep. Through 1976, samples had been collected down to 30 cm to measure total plutonium deposition and as a result of five years of sample collection and analysis at this depth, the total plutonium deposited in the Argonne environment has been established. By reducing the sampling depth to 5 cm, the analysis should be more sensitive to changes in current deposition. The grass samples were obtained by collecting the grass from a 1 m² area in the immediate vicinity of a soil sample. A grab sample technique was used to obtain bottom sediment. After drying, grinding, and mixing, 100 g portions of soil, bottom sediment, and grass were analyzed by the same method described in Section III.A. for air-filter residue. Results are given in terms of oven-dried soil, bottom sediment, or grass.

Comparison of the perimeter and off-site results in Table 12 shows that the same general range of concentrations exists in all areas for both plutonium isotopes, and it may be concluded that the plutonium in the perimeter samples resulted primarily from fallout of debris from nuclear detonations. Concentrations in the first 5 cm of soil are two to three times larger than in the first 30 cm. About one-third of the plutonium is in the top 5 cm.

Composite monthly precipitation samples were analyzed for plutonium-239. The results are given in Table 13, along with results since 1973 for comparison. The total 1979 deposition by precipitation was about a third that of 1978, a reflection of the decreased fallout from atmospheric nuclear tests, and was equivalent to 0.3% of the total plutonium previously deposited, which is reported to be 2.2×10^{-3} $\mu\text{Ci}/\text{m}^2$.⁽⁸⁾ The data in Table 13 is illustrated in Figure 5. The arrows at the bottom of the figure indicate the approximate dates of the Chinese atmospheric nuclear tests. Comparison of the results in Figure 5 with the plutonium air concentrations for the past seven years in Figure 4 shows excellent agreement. Cesium-137, radium-226, and thorium-228 were determined by gamma-ray spectrometry, while total uranium was determined

TABLE 12

Plutonium Content of Soil, 1979

Date Collected	Location	Plutonium-238		Plutonium-239		²³⁸ Pu/ ²³⁹ Pu
		10 ⁻⁹ μCi/g	10 ⁻³ μCi/m ²	10 ⁻⁹ μCi/g	10 ⁻³ μCi/m ²	
<u>Perimeter</u> *						
June 6	9N	0.9 ± 0.2	0.05 ± 0.01	9.8 ± 0.5	0.56 ± 0.03	0.093
June 6	13N	1.5 ± 0.2	0.09 ± 0.01	10.4 ± 0.5	0.63 ± 0.03	0.141
June 6	15H	2.5 ± 0.3	0.09 ± 0.01	40.0 ± 1.5	1.47 ± 0.06	0.063
June 6	13D	1.3 ± 0.2	0.07 ± 0.01	22.7 ± 0.9	1.28 ± 0.05	0.055
June 6	14H	0.8 ± 0.2	0.05 ± 0.01	16.8 ± 0.8	0.91 ± 0.04	0.050
December 10	8N	0.1 ± 0.2	< 0.01	2.6 ± 0.3	0.20 ± 0.02	0.015
December 10	10N	0.6 ± 0.2	0.05 ± 0.04	4.7 ± 0.6	0.40 ± 0.05	0.130
December 10	14L	1.4 ± 0.2	0.12 ± 0.02	19.3 ± 1.0	1.56 ± 0.08	0.074
December 10	12C	1.3 ± 0.3	0.08 ± 0.02	24.7 ± 1.2	1.43 ± 0.07	0.053
December 10	8G	0.9 ± 0.3	0.07 ± 0.02	20.1 ± 1.0	1.51 ± 0.07	0.044
	Average	1.1 ± 0.4	0.07 ± 0.02	17 ± 7	1.0 ± 0.3	0.072 ± 0.025
<u>Off-Site</u>						
June 5	Channahon, IL	1.5 ± 0.2	0.08 ± 0.01	15.2 ± 0.7	0.84 ± 0.04	0.097
June 5	Starved Rock State Park, IL	1.6 ± 0.2	0.10 ± 0.02	13.7 ± 0.7	0.84 ± 0.04	0.114
June 5	Morris, IL	1.4 ± 0.2	0.04 ± 0.01	28.1 ± 1.2	0.88 ± 0.04	0.048
June 12	Downers Grove, IL	0.4 ± 0.3	0.02 ± 0.01	11.1 ± 0.8	0.50 ± 0.03	0.031
June 12	Western Springs, IL	1.2 ± 0.2	0.07 ± 0.02	15.3 ± 0.8	0.97 ± 0.05	0.076
June 12	Brookfield, IL	0.6 ± 0.2	0.03 ± 0.01	17.4 ± 0.9	0.99 ± 0.05	0.035
October 4	Willow Springs, IL	0.7 ± 0.4	0.04 ± 0.02	16.3 ± 1.2	0.83 ± 0.06	0.045
October 4	Woodridge, IL	0.9 ± 0.3	0.04 ± 0.01	19.1 ± 1.0	0.88 ± 0.05	0.048
October 9	McKinley Woods State Park, IL	0.6 ± 0.3	0.04 ± 0.02	16.5 ± 0.6	0.96 ± 0.04	0.037
October 9	Dresden Lock and Dam, IL	0.3 ± 0.3	0.02 ± 0.02	6.6 ± 0.4	0.43 ± 0.03	0.045
	Average	0.9 ± 0.3	0.05 ± 0.02	16 ± 4	0.8 ± 0.2	0.058 ± 0.018

*The locations are given in terms of the grid coordinates in Figure 1.

TABLE 13

Ground Deposition of Plutonium-239,240
(Units of 10^{-6} $\mu\text{Ci}/\text{m}^2$)

Month	1973	1974	1975	1976	1977	1978	1979
January	-	0.60 ± 0.10	1.1 ± 0.18	0.25 ± 0.06	0.17 ± 0.08	0.37 ± 0.11	0.57 ± 0.20
February	0.14 ± 0.04	1.1 ± 0.16	1.7 ± 0.19	0.38 ± 0.09	0.15 ± 0.07	0.12 ± 0.09	0.43 ± 0.14
March	0.27 ± 0.06	2.0 ± 0.22	1.5 ± 0.17	0.58 ± 0.09	0.74 ± 0.15	4.7 ± 0.37	-
April	0.40 ± 0.07	5.2 ± 0.37	4.0 ± 0.43	0.21 ± 0.07	1.0 ± 0.17	3.3 ± 0.30	1.78 ± 0.25
May	0.50 ± 0.06	4.6 ± 0.31	1.8 ± 0.18	0.58 ± 0.11	1.8 ± 0.21	4.7 ± 0.42	0.58 ± 0.14
June	0.16 ± 0.04	3.5 ± 0.26	1.9 ± 0.18	0.65 ± 0.12	1.2 ± 0.18	3.0 ± 0.32	0.73 ± 0.17
July	0.32 ± 0.10	5.6 ± 0.37	0.48 ± 0.10	0.26 ± 0.07	0.71 ± 0.15	1.7 ± 0.24	0.45 ± 0.15
August	0.34 ± 0.14	1.0 ± 0.19	0.38 ± 0.09	0.15 ± 0.06	1.6 ± 0.21	0.54 ± 0.17	0.54 ± 0.14
September	0.27 ± 0.17	0.25 ± 0.11	0.10 ± 0.06	0.06 ± 0.05	1.5 ± 0.20	0.69 ± 0.18	0.21 ± 0.12
October	0.30 ± 0.12	0.45 ± 0.10	0.12 ± 0.08	0.30 ± 0.10	0.85 ± 0.15	0.34 ± 0.14	0.53 ± 0.22
November	0.22 ± 0.10	0.73 ± 0.12	0.08 ± 0.07	0.05 ± 0.04	0.72 ± 0.14	0.74 ± 0.16	0.09 ± 0.11
December	0.46 ± 0.12	0.71 ± 0.12	0.21 ± 0.08	0.12 ± 0.07	0.43 ± 0.10	0.40 ± 0.14	0.13 ± 0.10
Average Monthly Deposition	0.31 ± 0.07	2.1 ± 1.2	1.1 ± 0.7	0.30 ± 0.13	0.91 ± 0.31	1.7 ± 1.0	0.55 ± 0.27
Annual Deposition	3.38	25.74	13.37	3.59	10.87	20.60	6.04
Percent Added to Existing	0.2	1.2	0.6	0.2	0.5	0.9	0.3

PLUTONIUM-239, 240 GROUND DEPOSITION

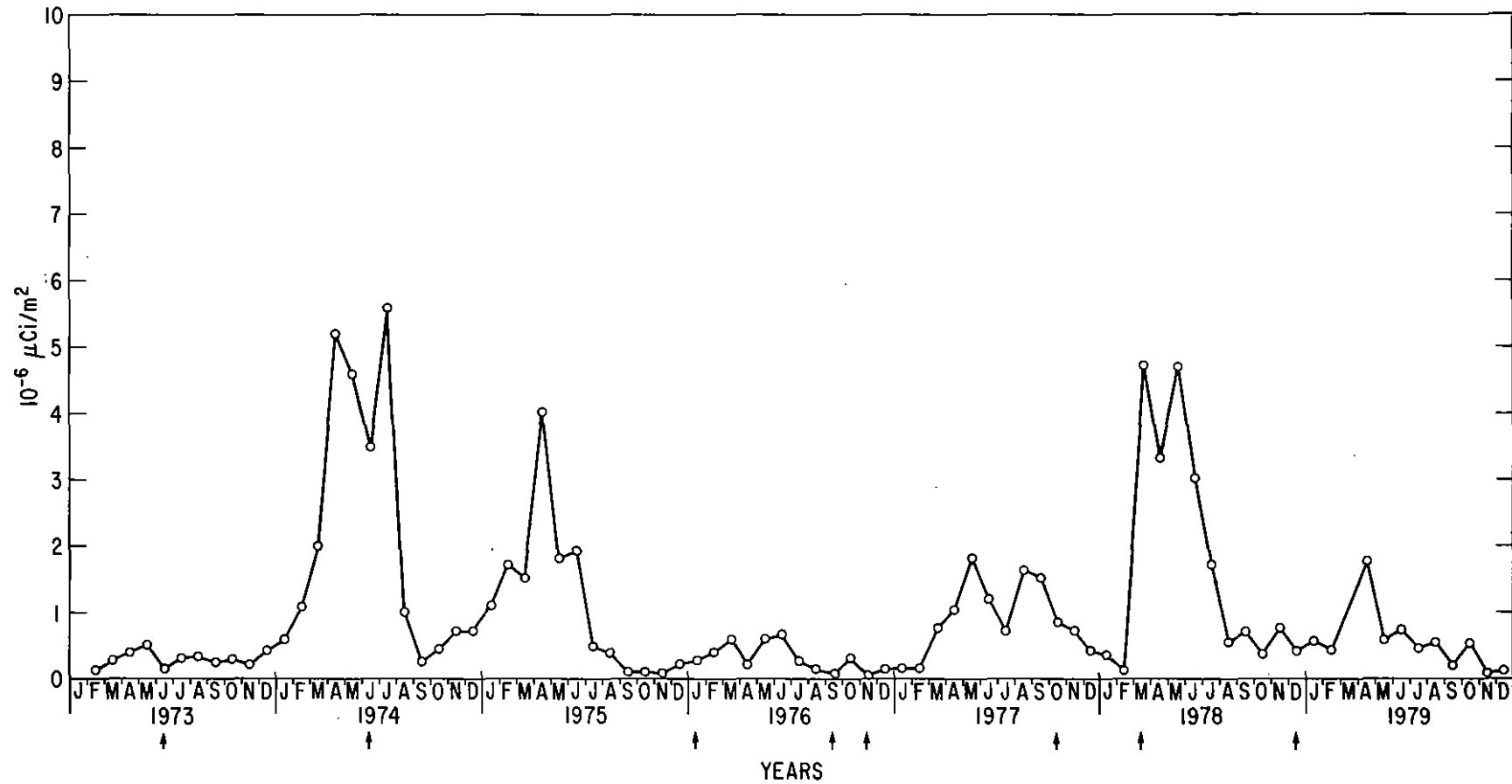


Fig. 5. Plutonium-239, 240 Surface Deposition, 1973-79

by photofluorometric analysis. All other activities were less than the detection limit of 10^{-7} $\mu\text{Ci/g}$. The concentrations in the perimeter and off-site samples, in Table 14, were similar and are the expected levels of the naturally-occurring radionuclides and fission products which are attributed to fallout. In terms of mass, the uranium concentrations are 4.2 $\mu\text{g/g}$ at the perimeter and 4.4 $\mu\text{g/g}$ off-site.

The sample collected at perimeter location 15H had the highest concentration of all of the radionuclides measured. The sample had a high organic content and was at one time on the bottom of a pond. It is our experience, based on similar samples collected in the past from off-site locations, that samples of this type of composition tend to have higher concentrations of radionuclides.⁽⁹⁾ The off-site samples collected at Morris, Illinois, had also in the past contained elevated concentrations of radionuclides. The source of this activity is at present undetermined.

The results of radioactivity measurements in grass are given in Tables 15 and 16. The grass samples were washed before analysis to remove surface soil, which contains considerably more radioactivity per gram than grass. If the grass is analyzed without washing, results are more variable and difficult to compare between locations. The perimeter concentrations are similar to results of previous years and within the expected range. Several off-site samples have elevated levels of various radionuclides, Starved Rock State Park, Illinois, Morris, Illinois, and Brookfield, Illinois, are probably due to incomplete removal of soil from the grass before analysis. This resulted in anomalously high average concentrations for the off-site samples. No fission product gamma-ray emitters were detected in any grass samples. In terms of deposition, the plutonium-239 concentration was a factor of about 10^4 less in the grass than in the soil from the same location.

Results of analysis of bottom sediment samples for cesium-137, radium-226, thorium-228, uranium, and plutonium are given in Table 17. Plutonium results vary widely between locations and are strongly dependent on the retentiveness of the bottom material. This is illustrated by the results from the sample collected from the bottom of the pond located at 15H. The composition of this sample was similar to the soil sample collected at the same location and to that of off-site bottom sediment samples collected at similar ponds in past years. The off-site concentrations were similar to results

TABLE 14

Radionuclides in Soil, 1979
(Concentrations in 10^{-6} $\mu\text{Ci/g}$)

Date Collected	Location	Cesium-137	Radium-226	Thorium-228	Uranium (natural)
<u>Perimeter</u> [*]					
June 6	9N	0.6 ± 0.1	1.2 ± 0.1	0.9 ± 0.1	1.4 ± 0.1
June 6	13N	0.5 ± 0.1	1.8 ± 0.2	0.9 ± 0.2	1.4 ± 0.2
June 6	15H	2.1 ± 0.1	1.2 ± 0.1	0.8 ± 0.1	1.8 ± 0.2
June 6	13D	1.3 ± 0.1	1.1 ± 0.1	0.9 ± 0.1	1.6 ± 0.2
June 6	14H	0.8 ± 0.1	1.1 ± 0.1	0.8 ± 0.1	1.3 ± 0.1
December 10	8N	0.1 ± 0.1	1.0 ± 0.1	0.6 ± 0.1	1.6 ± 0.1
December 10	10N	0.4 ± 0.1	1.0 ± 0.1	0.7 ± 0.1	1.5 ± 0.1
December 10	14L	1.1 ± 0.1	1.1 ± 0.1	0.7 ± 0.1	1.2 ± 0.1
December 10	12C	1.3 ± 0.1	0.8 ± 0.1	0.9 ± 0.1	1.1 ± 0.1
December 10	8G	1.1 ± 0.1	0.9 ± 0.1	0.8 ± 0.1	0.9 ± 0.1
	Average	0.9 ± 0.4	1.1 ± 0.2	0.8 ± 0.1	1.4 ± 0.2
<u>Off-Site</u>					
June 5	Channahon, IL	0.8 ± 0.1	0.9 ± 0.1	0.8 ± 0.1	1.1 ± 0.1
June 5	Starved Rock State Park, IL	0.7 ± 0.1	0.7 ± 0.1	0.4 ± 0.1	0.8 ± 0.1
June 5	Morris, IL	2.8 ± 0.1	2.3 ± 0.1	0.6 ± 0.1	3.8 ± 0.5
June 12	Downers Grove, IL	0.5 ± 0.1	1.2 ± 0.1	1.0 ± 0.1	1.0 ± 0.4
June 12	Western Springs, IL	0.8 ± 0.1	1.7 ± 0.1	0.9 ± 0.1	1.9 ± 0.2
June 12	Brookfield, IL	1.3 ± 0.1	1.3 ± 0.1	0.9 ± 0.1	1.6 ± 0.2
October 4	Willow Springs, IL	0.9 ± 0.1	1.2 ± 0.1	0.8 ± 0.1	0.9 ± 0.1
October 4	Woodridge, IL	1.1 ± 0.1	1.2 ± 0.1	0.8 ± 0.1	1.3 ± 0.1
October 9	McKinley Woods State Park, IL	1.0 ± 0.1	1.0 ± 0.1	0.8 ± 0.1	1.3 ± 0.1
October 9	Dresden Lock and Dam, IL	0.4 ± 0.1	1.2 ± 0.1	0.7 ± 0.1	1.4 ± 0.1
	Average	1.0 ± 0.4	1.3 ± 0.3	0.8 ± 0.1	1.5 ± 0.6

* The perimeter locations are given in terms of the grid coordinates in Figure 1.

TABLE 15

Plutonium Content of Grass Samples, 1979

Date Collected	Location	Plutonium-238		Plutonium-239	
		10^{-9} $\mu\text{Ci/g}$	10^{-6} $\mu\text{Ci/m}^2$	10^{-9} $\mu\text{Ci/g}$	10^{-6} $\mu\text{Ci/m}^2$
	<u>Perimeter</u> [*]				
June 6	9N	0.06 ± 0.10	0.015 ± 0.026	1.78 ± 0.27	0.445 ± 0.067
June 6	13N	0.10 ± 0.09	0.015 ± 0.015	0.52 ± 0.14	0.083 ± 0.023
June 6	15H	0.08 ± 0.12	0.008 ± 0.012	0.72 ± 0.19	0.073 ± 0.019
June 6	13D	0.02 ± 0.08	0.003 ± 0.019	0.97 ± 0.18	0.222 ± 0.042
June 6	14H	0.04 ± 0.01	0.005 ± 0.001	0.69 ± 0.20	0.087 ± 0.026
	Average	0.06 ± 0.03	0.015 ± 0.012	0.94 ± 0.45	0.180 ± 0.140
	<u>Off-Site</u>				
June 5	Starved Rock State Park, IL	0.37 ± 0.14	0.074 ± 0.028	5.13 ± 0.46	1.026 ± 0.092
June 5	Morris, IL	0.36 ± 0.16	0.115 ± 0.051	5.22 ± 0.50	1.670 ± 0.160
June 12	Downers Grove, IL	0.14 ± 0.02	0.016 ± 0.003	0.45 ± 0.16	0.052 ± 0.018
June 12	Western Springs, IL	0.01 ± 0.03	0.001 ± 0.003	1.25 ± 0.23	0.112 ± 0.021
June 12	Brookfield, IL	0.39 ± 0.22	0.020 ± 0.011	3.99 ± 0.51	0.200 ± 0.025
October 4	Willow Springs, IL	< 0.01	< 0.001	0.19 ± 0.12	0.027 ± 0.017
October 4	Woodridge, IL	< 0.01	< 0.001	0.15 ± 0.10	0.033 ± 0.022
October 9	McKinley Woods State Park, IL	< 0.01	< 0.001	0.49 ± 0.17	0.040 ± 0.014
October 9	Dresden Lock and Dam, IL	< 0.01	< 0.001	0.55 ± 0.20	0.080 ± 0.029
	Average	0.39 ± 0.12	0.115 ± 0.115	1.94 ± 1.46	0.359 ± 0.390

* The perimeter locations are given in terms of the grid coordinates in Figure 1.

TABLE 16

Radionuclides in Grass Samples, 1979
(Concentrations in 10^{-9} $\mu\text{Ci/g}$)

Date Collected	Location	Thorium-228	Thorium-230	Thorium-232	Uranium (natural)
<u>Perimeter</u> [*]					
June 6	9N	1.4 ± 0.4	2.9 ± 0.4	2.1 ± 0.3	27 ± 4
June 6	13N	1.3 ± 0.4	2.2 ± 0.4	1.5 ± 0.3	42 ± 5
June 6	15H	2.3 ± 0.7	2.6 ± 0.5	2.1 ± 0.4	24 ± 3
June 6	13D	1.6 ± 0.3	1.7 ± 0.3	1.6 ± 0.2	17 ± 3
June 6	14H	-	1.7 ± 0.6	0.8 ± 0.4	25 ± 3
	Average	1.7 ± 0.5	2.2 ± 0.5	1.6 ± 0.5	27 ± 8
<u>Off-Site</u>					
June 5	Starved Rock State Park, IL	-	-	-	23 ± 4
June 5	Morris, IL	2.3 ± 0.4	3.1 ± 0.3	2.2 ± 0.3	22 ± 3
June 12	Downers Grove, IL	2.5 ± 0.6	2.4 ± 0.4	2.0 ± 0.3	28 ± 3
June 12	Western Springs, IL	7.0 ± 1.1	11.2 ± 1.4	7.8 ± 1.0	72 ± 4
June 12	Brookfield, IL	12.8 ± 1.4	20.2 ± 1.9	13.3 ± 1.3	126 ± 9
October 4	Willow Springs, IL	2.4 ± 0.4	2.9 ± 0.4	2.3 ± 0.3	13 ± 4
October 4	Woodridge, IL	1.5 ± 0.3	1.8 ± 0.2	1.1 ± 0.2	32 ± 3
October 9	McKinley Woods State Park, IL	1.5 ± 0.1	1.8 ± 0.1	1.3 ± 0.1	56 ± 4
October 9	Dresden Lock and Dam, IL	4.0 ± 0.4	4.5 ± 0.3	3.4 ± 0.3	23 ± 3
	Average	4.3 ± 2.8	6.0 ± 4.6	4.2 ± 3.0	44 ± 24

*The perimeter locations are given in terms of the grid coordinates in Figure 1.

TABLE 17

Radionuclides in Bottom Sediment, 1979

Date Collected	Location	Concentrations in 10^{-6} $\mu\text{Ci/g}$				Concentrations in 10^{-9} $\mu\text{Ci/g}$	
		Cesium-137	Radium-226	Thorium-228	Uranium (natural)	Plutonium-238	Plutonium-239
	<u>Perimeter</u> *						
June 6	15H	2.0 ± 0.4	0.8 ± 0.2	0.7 ± 0.2	1.6 ± 0.1	2.1 ± 0.3	36.3 ± 1.8
	<u>Off-Site</u>						
June 5	DuPage River, Channahon, IL	0.3 ± 0.1	1.0 ± 0.3	1.2 ± 0.3	1.2 ± 0.1	1.0 ± 0.2	6.2 ± 0.4
June 12	Salt Creek, Western Springs, IL	0.1 ± 0.1	1.3 ± 0.4	0.7 ± 0.2	1.5 ± 0.1	0.5 ± 0.1	1.3 ± 0.2
June 12	Des Plaines River, Brookfield, IL	0.4 ± 0.1	0.6 ± 0.2	0.5 ± 0.2	1.0 ± 0.1	0.6 ± 0.1	7.6 ± 0.5
October 4	Flagg Creek, Willow Springs, IL	0.2 ± 0.1	1.2 ± 0.3	0.7 ± 0.2	0.8 ± 0.1	1.0 ± 0.3	4.5 ± 0.4
October 4	DuPage River, West Chicago, IL	0.1 ± 0.1	1.0 ± 0.3	0.8 ± 0.2	1.2 ± 0.1	0.6 ± 0.2	2.4 ± 0.3
October 4	DuPage River, Naperville, IL	0.4 ± 0.1	0.8 ± 0.2	1.3 ± 0.4	1.1 ± 0.1	0.3 ± 0.4	10.6 ± 0.6
October 9	Illinois River, Starved Rock State Park, IL	< 0.1	0.2 ± 0.1	0.1 ± 0.1	0.2 ± 0.1	< 0.1	0.5 ± 0.1
October 9	Illinois River, Morris, IL	< 0.1	0.3 ± 0.1	0.2 ± 0.1	0.5 ± 0.1	< 0.1	0.2 ± 0.1
	Average	0.2 ± 0.1	0.8 ± 0.3	0.7 ± 0.3	0.9 ± 0.3	0.5 ± 0.3	4.2 ± 2.6

* The perimeter location is given in terms of the grid coordinates in Figure 1.

from previous years. The other radionuclide concentrations are normal and similar to those found in soil. In terms of mass, the uranium concentrations averaged 2.7 $\mu\text{g/g}$ at the off-site locations.

4. Foodstuffs

Raw milk was collected monthly from a local dairy farm south of Lemont and analyzed for several radioactive nuclides by methods similar to those used for water. Iodine-131 was analyzed with a detection limit of 1×10^{-10} $\mu\text{Ci/ml}$ by an ion-exchange separation followed by beta counting. Cesium-137 was analyzed with a detection limit of 5×10^{-10} $\mu\text{Ci/ml}$ by an ion-exchange separation followed by gamma-ray spectrometry. The other nuclides were analyzed by the same methods used for water and with the same detection limits. The results are given in Table 18. The average strontium-90 concentration decreased by about 15% compared to 1978, while the average cesium-137 concentration decreased by 25%. These nuclides are fission products from nuclear tests and their presence in milk is not related to Argonne operations. No short-lived fission products, i.e., strontium-89, iodine-131, or barium-140, were detected in milk.

The concentrations given in Table 18 may be compared to the CGs for drinking water given in the Appendix, Part B. The drinking water CGs are based on an intake of 2.2 liters per day. The consumption of one liter of milk per day would result in an average intake of 0.55% of the strontium-90 and 0.006% of the cesium-137 Concentration Guides.

Several samples of garden vegetables grown 8-16 km (5-10 miles) from Argonne were analyzed for plutonium, uranium, and thorium. The results are given in Table 19, and the concentrations are expressed in terms of air-dried weight. Compared to the results for grass, the plutonium-239 concentrations are about a hundred times lower, while the uranium concentrations are similar. The uranium results are in good agreement with analyses of the same types of vegetables measured during the past several years. The plutonium content, although extremely variable, covered about the same range of concentrations as in the last three years. Included with the plutonium results is a concentration factor (CF), which is defined as the ratio of plutonium concentration in the food divided by the average plutonium concentration in soil. As in the case of milk, the radioactivity is unrelated to Argonne operations, but the information is valuable as background data.

TABLE 18

Radionuclides in Milk, 1979
(Concentrations in 10^{-9} $\mu\text{Ci/ml}$)

Date Collected	Hydrogen-3	Strontium-89	Strontium-90	Iodine-131	Cesium-137	Barium-140
January 10	< 100	< 2	3.2	< 0.1	3.3	< 2
February 7	< 100	< 2	4.2	< 0.1	5.1	< 2
March 7	110	< 2	2.5	< 0.1	4.3	< 2
April 4	< 100	< 2	3.2	< 0.1	4.3	< 2
May 2	< 100	< 2	3.5	< 0.1	4.5	< 2
June 6	159	< 2	4.8	< 0.1	3.1	< 2
July 3	-	< 2	5.2	< 0.1	1.8	< 2
August 1	< 100	< 2	3.3	< 0.1	1.3	< 2
September 6	126	< 2	4.6	< 0.1	1.2	< 2
October 3	< 100	< 2	4.0	< 0.1	0.9	< 2
November 7	< 100	< 2	3.2	< 0.1	1.4	< 2
December 5	< 100	< 2	2.6	< 0.1	1.2	< 2
Average	< 109	< 2	3.7 ± 0.5	< 0.1	2.7 ± 0.9	< 2

TABLE 19

Radionuclides in Garden Vegetables, 1979

Food	Concentration (10^{-9} $\mu\text{Ci/g}$)				Plutonium-239	
	Thorium-228	Thorium-230	Thorium-232	Uranium (natural)	Concentration (10^{-12} $\mu\text{Ci/g}$)	CF
Tomatoes	< 0.1	< 0.1	< 0.1	32 ± 4	< 1	-
Cabbage	< 0.1	< 0.1	< 0.1	11 ± 3	< 1	-
Corn (kernels)	< 0.1	< 0.1	< 0.1	22 ± 1	< 1	-
Swiss Chard	3.3 ± 0.5	5.6 ± 0.5	3.2 ± 0.4	61 ± 6	16 ± 9	8×10^{-4}

5. External Penetrating Radiation

Measurements were made with calcium fluoride (dysprosium activated) and lithium fluoride thermoluminescent dosimeter (TLD) chips. Each measurement was the average of three or four chips exposed in the same packet. The response of the chips was calibrated with an NBS standard radium-226 source, and the results calculated in terms of air dose. Dosimeters were exposed at a number of locations at the site boundary to determine any dose that might be due to Argonne operations at the closest uncontrolled approaches to the Laboratory, and at several locations on the site. The latter were chosen for two purposes; to determine where abnormal doses might be encountered, and where the results might be useful in determining the origin of any abnormal dose readings obtained at the boundary. Readings were also taken at five off-site locations for comparison purposes. These locations are shown in Figure 2.

The results are summarized in Tables 20 and 21, and the site boundary and on-site readings are also shown in Figure 6. Measurements were made in six successive exposure periods that varied in length from 54 to 71 days, and in total covered the period from January 9, 1979, to January 16, 1980. The results for each period were calculated in terms of annual dose for ease in comparing measurements made for different elapsed times, and were weighted according to their exposure times in calculating the annual average at each location. The error given in the tables for an average is the 95% confidence limit calculated from the standard error.

The off-site results averaged 89 ± 7 mrem/yr. The standard error of the mean of all 30 individual results was 2.0 mrem/yr. The standard deviation of the average of the five separate locations was 7 mrem/yr. From 1973 to 1978, the corresponding averages and standard errors varied from 90 ± 2 mrem/yr (1976) to 100 ± 2 mrem/yr (1973). The six-year average was 95 ± 4 mrem/yr. Thus, the background radiation has been quite consistent and the 1979 average is within the range found previously. The variations from year-to-year have also been consistent at each location. The annual average at Downers Grove, Lemont (previously given as Lockport), and Lombard have been within 5 mrem/yr of the average of all five off-site locations (usually within 2 mrem/yr). At Oak Lawn, the results have been from 6 to 11 mrem/yr below, and at Oakbrook, 6 to 11 mrem/yr above, the overall average.

TABLE 20

Environmental Penetrating Radiation at Off-Site Locations, 1979

Location	Dose Rate (mrem/year)						Average
	Period of Measurement						
	1/9-3/15	3/15-5/15	5/15-7/10	7/10-9/13	9/13-11/6	11/6-1/16	
Downers Grove	68	89	95	84	92	90	86 ± 10
Lemont	71	91	98	95	98	96	91 ± 10
Lombard	69	89	98	88	97	90	88 ± 10
Oak Lawn	70	84	85	82	80	84	81 ± 6
Oakbrook	77	105	100	104	109	101	99 ± 11
Average	71 ± 4	92 ± 9	95 ± 7	91 ± 10	95 ± 12	92 ± 7	89 ± 7

TABLE 21

Environmental Penetrating Radiation at ANL, 1979

Location	Dose Rate (mrem/year)						Average
	Period of Measurement						
	1/9-3/15	3/15-5/15	5/15-7/10	7/10-9/13	9/13-11/6	11/6-1/16	
14L - Boundary	64	82	81	82	87	82	80 ± 8
14I - Boundary	118	148	148	143	153	153	144 ± 17
14G - Boundary	57	-	87	90	92	87	82 ± 16
13D - Boundary	42	60	62	64	66	66	60 ± 9
9/10EF - Boundary	56	85	93	91	89	-	83 ± 17
8H - Boundary	63	88	91	86	94	90	85 ± 11
8H - Center, St. Patrick's Cemetery	73	103	105	100	118	106	101 ± 15
7I - Boundary	405	411	417	410	480	559	447 ± 61
6I - 200 m N of Quarry Road	77	89	96	91	108	105	94 ± 11
9L - Boundary	56	70	71	67	74	69	68 ± 6
9H - 50 m SE of CP-5	2100	2440	2120	1890	1930	1940	2070 ± 210
8H - 65 m S of 316	92	96	107	105	121	106	104 ± 10
9H - 23 m E of 316	166	96	86	-	-	-	118 ± 44
9I - 45 m NE of 350 210 m NE of 316	94	95	94	94	-	-	94 ± 1
9I - 65 m NE of 350 235 m NE of 316	-	-	-	-	85	79	82 ± 6
8H - 200 m NW of Waste Storage Area (Heliport)	118	157	155	154	169	172	154 ± 19
7I - Center, Waste Storage Area	7440	7320	7080	7200	-	3120	6340 ± 2130
10/11K - Lodging Facilities	62	70	68	68	80	76	71 ± 6
9J - Between ZGS Condenser and 386	277	312	268	219	211	165	240 ± 53
13J - 135 m E of 202	253	250	245	243	258	252	250 ± 5
12M - 30 m W of 55	131	69	80	66	62	63	79 ± 27

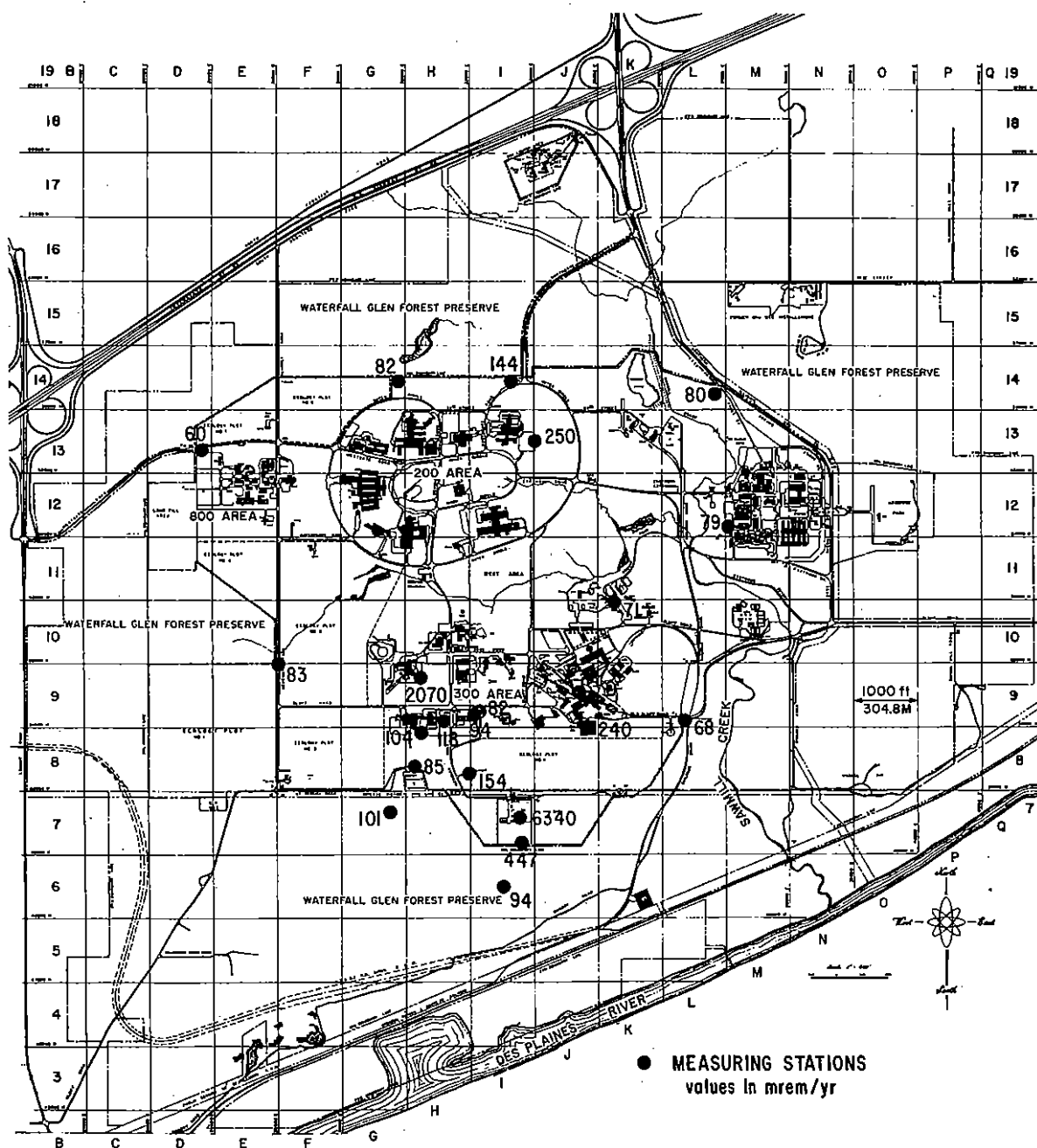


Fig. 6. Penetrating Radiation Measurements at the ANL Site, 1979

If the off-site locations are an accurate sample of the radiation background in the area, then annual averages at the site boundary in the range of 89 ± 7 mrem/yr may be considered normal with a 95% probability, or 95% of the boundary results should have averages in this range. To compare boundary results for individual sampling periods, the standard deviation of the mean of the 30 individual off-site results is useful. This value is 11 mrem/yr, so that individual results in the range of 89 ± 22 mrem/yr may be considered normal with a 95% probability.

At two locations, 7I at the south boundary of the site, and at 14I north of Building 202, the dose rates were consistently above the normal range. At 7I, this was due to radiation from a Radioactive Waste Storage Facility in the northern half of grid 7I. Waste is packaged and temporarily kept in this area prior to removal for permanent storage elsewhere. The net above-normal dose at this location was about 360 mrem/yr; in previous years this value has ranged from 440 mrem/yr in 1976 to 114 mrem/yr in 1977. The dose at the 7I boundary from the Waste Storage Facility is consistent with the dose at the center of the Facility; the boundary dose is about 5% of the dose measured at the center.

At 14I, the average dose rate was about 55 mrem/yr above normal, the same as the 1978 rate. This dose is attributed to the use of cobalt-60 irradiation sources in Building 202. Of interest is the fact that the dose rates at 14I and at all off-site locations were significantly lower during the first measurement period, when the snow cover was heavy and temperatures low, but the difference during the first period (46 mrem/yr) was similar to the average difference for the year (55 mrem/yr). The dose at 14I responds to changes in the frequency of use of the sources, so the rate is not expected to remain constant during all measurement periods.

Dose variations from period to period at the same location, which are at least in part statistical, and natural dose differences between locations make it difficult to determine with high certainty when site boundary doses are only a few mrem/yr above normal and due to Argonne operations. Three criteria are used to identify such locations: 1) the results for each sampling period are frequently above the off-site average for the same period; 2) the annual average at a location exceeds the off-site average (89 mrem/yr) plus the standard deviation in the average at the 95% confidence limit (7 mrem/yr);

or 3) occasional results at a particular location are significantly above the normal value for that location, but do not exceed the off-site normal range. The last criterion could apply to a location such as 13D, where the results have been consistently below the average. At this location, the ground contains considerable gravel, which has less radium than the usual clay soil, and this probably accounts for the low dose rates. Application of the first two criteria and the difficulties in interpreting small differences from natural background by TLD measurements are illustrated below.

The dose rate at 8H is of interest, since it lies in St. Patrick's Cemetery, which was in use before Argonne was constructed, and which is open to visitors. The dose at the center of the Cemetery was 12 mrem/yr above the off-site average, but 5 mrem/yr above the upper limit (96 mrem/yr) of the normal range. The result for one individual period (118 mrem/yr) was above the 95% upper limit for such measurements, 111 mrem/yr. The doses for individual measurement periods exceeded the off-site averages for the same periods by 2 mrem/yr to 23 mrem/yr. Based on these types of comparison, the above-normal dose at 8H is estimated to be 5 to 10 mrem/yr. Similar results were obtained in previous years.

At the south end of the Laboratory site, three possible sources of external radiation exist; direct radiation from the Waste Storage Facility, direct radiation from a tandem dynamitron and low-power reactors in Building 316 (location 9GH), and argon-41 from the CP-5 reactor at location 9H. The contribution from CP-5 and 316 to the dose at 8H is considered negligible since dose rates measured in other directions from these sources at the same distance were less than at 8H. In addition, the dose at the Cemetery did not decrease when the dose close to Building 316 decreased after the reactor in that building was provided with additional shielding, and the dose at the center of the Cemetery averaged 16 mrem/yr above that at the 8H boundary station, which is closer to Building 316. For these reasons, the excess dose is attributed to the material stored at 7I, rather than to sources north of the Cemetery.

6. Potential Radiation Dose Estimates

a. Air and Waterborne Radionuclides

The radiation doses at the site boundary and off the site that could have been received by the public from radioactive materials leaving the site were calculated by two methods. Where measured radionuclide concentrations in air and water are available, conversion of concentration to dose was based on the ratio of environmental concentrations to the Concentration Guides given in Table 22. This table gives the annual radiation doses that would result from continuous exposure at the specified concentrations for those nuclides whose presence in the environment are attributable to Argonne. For argon-41, tritiated water vapor, and iodine-131 released from reactor stacks, doses were calculated from an atmospheric dispersion model which made use of a source term and meteorological data.

The principal exposure pathway for radioactive substances released from Argonne is directly from air to man by inhalation. Although Sawmill Creek water is not used for drinking purposes, the dose that would be received by an individual ingesting water at the concentrations found in the Creek is calculated. A minor exposure route is from water to man by ingestion of water 711 km (370 miles) downstream from Argonne (Section I.E.), but the dilution of Sawmill Creek water at this point is so great that the dose calculation is meaningless. No other exposure pathways are significant.

Argon-41 and hydrogen-3 (in the form of tritiated water) from the CP-5 reactor represent the major portion of the gaseous radioactive effluent released from the Laboratory. The concentrations and dose rates, as a function of distance from CP-5, were calculated for these two nuclides by a computer program based on an atmospheric dispersion model.⁽¹¹⁾ The following parameters were used in the calculations:

- a) release rates (measured in the CP-5 exhaust stack): argon-41, 1.03 Ci/MW-hr; hydrogen-3, 0.076 Ci/hr.
- b) meteorological data: the wind velocity data shown in Figure 3.
- c) the usual parameters for stack height, building, wake, plume, momentum, temperatures, etc.

The calculations were carried out to 80 km (50 miles). The argon-41 results for the first 4.8 km (3 miles) are given in Table 23. Doses were

TABLE 22

Concentration-to-Dose Conversion Factors

Nuclide	Medium	Concentration* ($\mu\text{Ci/ml}$)	Dose* (rem)	Critical Organ
Americium-241	Water	4×10^{-6}	1.5	Kidney
	Water	5×10^{-6}	3	Bone
Argon-41	Air	4×10^{-8}	0.5	Whole Body
Californium-249	Water	4×10^{-6}	3	Bone
Californium-252	Water	7×10^{-6}	1.5	GI (LLI)
	Water	2×10^{-5}	3	Bone
Curium-242	Water	2×10^{-5}	1.5	GI (LLI)
	Water	2×10^{-4}	3	Bone
Curium-244	Water	7×10^{-6}	3	Bone
Hydrogen-3 (H_2O)	Air	2×10^{-7}	0.5	Whole Body
	Water	3×10^{-3}	0.5	Whole Body
Iodine-131	Air	1×10^{-10}	1.5	Thyroid
Neptunium-237	Water	3×10^{-6}	3	Bone
Plutonium-238	Water	5×10^{-6}	3	Bone
Plutonium-239	Water	5×10^{-6}	3	Bone
Strontium-90	Water	3×10^{-7}	3	Bone

*The concentrations and doses are the Radiation Protection Standards (RPS) specified in DOE Manual Chapter 0524 for individuals in uncontrolled areas, except for americium-241 (bone), californium-252 (GI and bone), and curium-242 (bone). Since RPS values for these nuclide-organ combinations are not given in DOE Manual 0524, the concentrations used were one-tenth of the 168 hour occupational values specified by the ICRP. (10)

TABLE 23

Argon-41 Radiation Dose From CP-5 Reactor, 1979
(millirem/year)

Sector	Distance			
	1.5 km (0.93 mi)	0-1.6 km (0-1 mi)	1.6-3.2 km (1-2 mi)	3.2-4.8 km (2-3 mi)
N	1.1	2.7	0.5	0.2
NNE	1.3	3.2	0.6	0.3
NE	1.0	2.5	0.5	0.2
ENE	1.1	2.7	0.5	0.2
E	0.8	2.1	0.4	0.2
ESE	0.8	2.0	0.4	0.2
SE	0.8	1.9	0.4	0.2
SSE	0.7	1.6	0.3	0.2
S	0.7	1.7	0.3	0.2
SSW	0.8	2.0	0.4	0.2
SW	0.7	1.7	0.3	0.2
WSW	0.6	1.5	0.3	0.1
W	0.4	1.1	0.2	0.1
WNW	0.5	1.2	0.3	0.1
NW	0.6	1.4	0.3	0.1
NNW	0.7	1.7	0.3	0.2
Average	0.8	1.9	0.4	0.2

calculated for the mid-point of the annular interval. Thus, the dose for the 0-1.6 km (0-1 mile) average is the dose at 0.8 km (0.5 mile). The highest dose rates are in the N to E sectors. In this area, the full-time residents who receive the largest dose, 0.6 mrem/yr (outdoors) live 2.2 km (1.4 miles) from the reactor in the NE direction. This dose is less than 0.2% of the standard (500 mrem/yr) for individuals in uncontrolled areas. The dose varies greatly with distance in the first several kilometers. Thus, in the NE direction, individuals would receive 1.2 mrem/yr if they were outdoors throughout the year at 1.6 km (1 mile) and 0.2 mrem/yr if they were outdoors at 3.2 km (2 miles).

The measurement technique for argon-41 is adequate in the vicinity of CP-5, but is not sufficiently sensitive to measure the concentration at the site boundary. However, an upper limit for the argon-41 dose at the site boundary can be estimated from the penetrating radiation dose measurements made with thermoluminescent dosimeters (TLD) and discussed in Section III.A.1. The measurements made south of the reactor and north of Building 202 are obscured by direct radiation from several gamma-ray sources, but in other directions, including the predominant wind directions to the east and northeast, the dose rates at the site boundary were in the normal range found off-site. Increases in excess of two standard deviations of any single off-site result (i.e., greater than 89 ± 22 mrem/yr) would have been recognized as abnormal, and on this basis, the dose from argon-41 at the site boundary was less than about 22 mrem/yr. Thus, the calculated doses and those measured by TLD agree within the ability of the TLD system to detect above-normal doses.

The population data in Table 2 was used to calculate the population dose from argon-41. The results are given in Table 24, together with the average individual dose. For comparison, the table also gives the estimated natural external radiation dose, which was calculated with the assumption that the average off-site, outdoor radiation dose measured by TLD applies to the entire area within an 80 km (50 miles) radius.

The dose rates due to hydrogen-3 from CP-5, calculated in the same manner as for argon-41, are as follows. The maximum dose (outdoors) at 1.5 km (0.93 mile), in the NNE direction, is 0.020 mrem/yr. The maximum individual exposure to full-time residents in the area occurs in the 1.6-3.2 km (1-2 miles) annulus in the NNE direction, where the annual dose is calculated to be

0.013 mrem/yr. A summary of the results is given in Table 25. A comparison of the doses calculated from the meteorological model with the measured data from Table 8 is given in Table 26. In both cases, concentrations are converted to dose as described earlier. The agreement is good in view of the large number of variables and parameters involved in obtaining both the calculated and measured values.

TABLE 24

Argon-41 Average Individual and Population
Dose From CP-5 Reactor, 1979

Distance (km)	Population	Dose			
		Avg. mrem/year		Man-rem/year	
		Argon-41	Natural	Argon-41	Natural
0-1.6	0	-	-	-	-
1.6-3.2	5,559	0.4	89	2.2	495
3.2-4.8	17,692	0.2	89	3.5	1575
0-80	7,941,000	0.0048	89	38	7.07×10^5

TABLE 25

Hydrogen-3 Average Individual and Population
Dose From CP-5 Reactor, 1979

Distance (km)	Population	Avg. mrem/year	Man-rem/year
0-1.6	0	-	-
1.6-3.2	5,559	0.0049	0.027
3.2-4.8	17,692	0.0033	0.058
0-80	7,941,000	0.00026	2.04

TABLE 26

Comparison of Calculated and Measured
Hydrogen-3 Dose Rates, 1979

Direction	Distance (km)	Calculated (mrem/yr)	Measured (mrem/yr)
NNE	1.5	0.018	0.020
ENE	1.9	0.011	0.012
SW	0.45	0.055	0.032

The iodine-131 released from CP-5 would result in an individual dose of 0.002 mrem/yr at 1.5 km (0.93 mile) in the NNE sector and a population dose of about 0.001 man-rem/yr in the 1.6-3.2 km (1-2 miles) annulus. These values are so small that the calculations for greater distances are not worthwhile.

The only location where radionuclides attributable to Argonne operations could be found in off-site water was Sawmill Creek below the waste-water outfall. The nuclides added to Sawmill Creek by Argonne waste water, their net concentrations in the Creek, and the corresponding dose rates if water at these concentrations were used as the sole water supply by an individual are given in Table 27. The dose rates were all well below the standards for individuals in uncontrolled areas. It should be emphasized that Sawmill Creek is not used for drinking, swimming, or boating. Inspection of the area shows there are few fish in the stream, and they do not constitute a significant source of food for any individual.

TABLE 27

Radionuclide Concentrations and Dose Estimates
for Sawmill Creek Water, 1979

Nuclide	Conc. (net avg.) 10^{-9} μ Ci/ml	Dose mrem/year	Percent of Standard
Hydrogen-3	242	0.040	0.008
Strontium-90	0.04	0.4	0.013
Neptunium-237	0.0055	0.0055	0.0002
Plutonium-238	< 0.0015	< 0.0009	< 0.00003
Plutonium-239	0.015	0.009	0.0003
Americium-241	0.0098	0.0037 (kidney)	0.00024
		0.0059 (bone)	0.00020

As indicated in Table 9, occasional Creek samples (less than 10) contained traces of curium and californium nuclides, but the averages were only slightly greater than the detection limit. The annual dose due to an individual consuming water at these concentrations can be calculated as was done for those nuclides more commonly found in Creek water, but it should be noted that the method of averaging probably exaggerates the true concentration.

These annual doses range from 2×10^{-4} to 5×10^{-5} mrem/yr for these nuclides.

The EPA⁽¹²⁾ has recently established drinking water standards based on a dose of 4 mrem/yr. The standards in Table 22 are based on doses of 500 mrem/yr to 3000 mrem/yr, depending on the critical organ. Except for strontium-90, the percent of the EPA standard can be obtained by multiplying the value in Table 27 by the ratio 500/4 for hydrogen-3 and 3000 or 1500/4 (depending on the critical organ) for the actinide elements. Thus, for hydrogen-3, the value becomes 1.0%. For strontium-90, the EPA, using a different model than reference (10), developed a standard of 8×10^{-9} $\mu\text{Ci/ml}$ to produce a dose of 4 mrem/yr and therefore the concentration in Table 27 would give 0.5% of that standard.

b. External Penetrating Radiation

Above normal fence-line doses attributable to Argonne operations were found at the south boundary near the Waste Storage Facility (location 7I), at the north boundary near Building 202 (14I), and probably at the south boundary adjacent to St. Patrick's Cemetery (8H). The results are discussed in Section III.A.5.

At location 7I, the fence-line dose from Argonne was about 360 mrem/yr. Approximately 300 m (0.3 mile) south of the fence line (grid 6I), the measured dose of 94 ± 11 mrem/yr was within the normal range. There are no individuals living in this area. The closest residents are about 1.6 km (1 mile) south of the fence line. At this distance, the calculated dose rate from the Waste Storage Area (based on exponential absorption of the radiation, a decrease in intensity with the square of the distance, and an increase in intensity with distance due to the buildup factor) is 0.01 mrem/yr, if the energy of the radiation was 0.66 MeV, and 0.04 mrem/yr, if the energy was 1.3 MeV. The energy spectrum of the radiation varies with time and is not known, so it is necessary to assume an energy to make the calculations. Since cesium-137 and cobalt-60 are common radionuclides, the energies of the gamma-rays from these nuclides were used in the calculations.

At St. Patrick's Cemetery (8H), the upper limit of the estimated dose attributable to Argonne was estimated at 5-10 mrem/yr. An individual spending an average of 1 hr/week at this location would receive an annual dose in the range of 0.003 mrem/yr to 0.006 mrem/yr.

In the area north of the site, where the fence-line radiation dose from the cobalt-60 sources in Building 202 was measured at about 55 mrem/yr, the nearest residents are 750 m (0.47 mile) to the north-northwest. The dose at that location (calculated as described above) was about 0.1 mrem/yr. In addition, the argon-41 dose here is about 0.5 mrem/yr.

The applicable Radiation Protection Standards for whole body external radiation dose to the general population is a maximum of 500 mrem/yr to critical individuals, or if individual doses are not known, 170 mrem/yr to a suitable sample of the exposed population.⁽⁴⁾ The latter criterion assumes that the maximum dose to individuals in the sample will not exceed the average by more than a factor of three. Thus, the doses to individuals living near the site will not exceed 0.1% of the 500 mrem/yr limit or 0.3% of the "suitable sample" limit. Argon-41 constitutes about 98% of this dose. At the fence line, where higher doses were measured, the land is wooded and unoccupied.

All of these dose calculations are based on full-time, outdoor exposure. Actual exposures to individuals are substantially less since they are inside (which provides shielding) or away from their dwelling much of the time.

c. Summary

The total dose received by off-site residents was combined from the separate pathways that contribute to this total: argon-41 immersion dose, hydrogen-3 inhalation dose, and cobalt-60 external radiation dose. The highest dose was about 0.6 mrem/yr (essentially from argon-41) to individuals living NE of the site if they were outdoors at that location during the entire year. The total annual population dose to the entire area within an 80 km (50 mile) radius is about 40 man-rem.

B. Chemical and Biological Pollutants

1. Water

The environmental water data contained in this section have been collected in an effort to ascertain Argonne compliance with State of Illinois regulations on surface stream and effluent water quality, as well as to verify the adequacy of Argonne's effluent pollution controls. The appropriate standards are listed in the Illinois Pollution Control Board Rules and Regulations, Chapter 3.⁽¹³⁾

Stream Quality Standards appear in Part II and Effluent Quality Standards appear in Part IV of that document.

The flameless atomic absorption technique using pyrolytic graphite has been extended to barium and total chromium. This technique is also the standard procedure for cadmium, copper, lead, manganese, nickel, and silver. The lower limits of detection for all of these elements have been significantly reduced. The x-ray fluorescence technique has been applied to selenium as well as to arsenic. Selenium results appear for the first time.

The results of the measurement of chemical constituents are expressed as milligrams (mg) or micrograms (μ g) per liter (l). Averages were calculated as described in Section III.A. Yearly averages are reported with a (\pm) limit value. This value is the standard error at the 95% confidence limit and it is calculated from the standard deviation of the yearly average. Only when sample concentrations are random does this value approach the actual distribution occurring at the sampling location. In some instances, it appears that the measurements do represent a natural background concentration and the variation is representative of climatic conditions.

All of the results are compared to the appropriate State standards, which are listed in Table 28. Minimum detectable amounts are included for comparison. The detection limits for the atomic absorption methods represent twice the background variation, which is commonly used for this purpose. Detection limits for ion selective methods are those listed by the manufacturer. The lower limit of detection for x-ray fluorescence is based on the appropriate counting statistics.

As in the past, emphasis has been placed on three areas: Sawmill Creek, a tributary of the Des Plaines River, since this is the principal route for waste water leaving the Argonne site; the study of cooling tower blowdown effluents; and the control of mercury release in the effluent.

a. Effluent Water

The major discharge of waste water from Argonne operations is by way of the waste treatment plant. The water volume from this source was approximately 2.7 megaliters (0.73 million gallons) per day and in 1979 was comprised of 35% sanitary waste water and 65% water from laboratory operations. The laboratory waste water is held in 0.26 megaliter (69,000 gallon) tanks and

TABLE 28

Water Quality Standards and Detection Limits

(Concentrations in mg/l)

Constituent	State Standard		Analytical Detection Limit
	Stream	Effluent	
Ammonia Nitrogen (as N)	1.5	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	0.1
Arsenic	1.0	0.25	0.01
Barium	5.0	2.0	0.005
Cadmium	0.05	0.15	0.0004
Chromium (hexavalent)	0.05	0.3	0.01
Chromium (trivalent)	1.00	1.00	-
Copper	0.02	1.0	0.0008
Cyanide	0.025	0.025	0.020
Fluoride	1.4	15	0.02
Iron	1.0	2.0	0.05
Lead	0.1	0.1	0.002
Manganese	1.0	1.0	0.001
Mercury	0.0005	0.0005	0.0001
Nickel	1.0	1.0	0.003
pH	6.5-9.0	5.0-10.0	-
Selenium	1.0	1.0	0.01
Silver	0.005	0.1	0.0002
Sulfate	500	-	1.0
Temperature	-	-	0.1°C
Total Dissolved Solids	1000.	-	-
Zinc	1.0	1.0	0.01

is checked for radioactivity before release. The release of these tanks occurs with some periodicity at a rate of about 4,200 liters (1,100 gallons) per minute. The sanitary waste water is released at a reasonably constant rate during the entire 24-hour period.

The performance of the sanitary waste treatment plant was monitored by the Reclamation Control Laboratory of the Plant Systems Division. This was done by analyzing twice weekly samples of the combined sanitary and laboratory waste systems for biochemical oxygen demand (B.O.D.), suspended solids, and ammonia nitrogen content. Each sample was a composite of eight separate grab samples taken approximately once per hour. All analyses were performed as outlines in Standard Methods.⁽¹⁴⁾

Release of chemical pollutants from the waste treatment plant was monitored on a continuous basis during the work week. A flow proportional 24-hour sample of the combined sanitary and laboratory effluent was obtained each day and was analyzed for constituents of interest.

Waste water at 11L was diverted in 1978 and this was described in the 1978 report.⁽¹⁵⁾ This channel is frequently dry, but sampling has been performed periodically to evaluate any residual hexavalent chromium and silver which might still be present.

In 1979, blowdown water from buildings in the 200 Area was combined and the effluent from this combination is monitored at 12L. Sampling at this location was either once per week grab sampling or continuous monitoring. Grab samples were taken when mechanical difficulties precluded use of the continuous monitoring equipment. Monitoring continued on a once per month basis, when possible, at location 12F, 14G, 14H, and 14J to determine whether any residual chromium exists. Use of non-chromate corrosion inhibitors was instituted extensively during this period. Since these substitutes contain zinc, this element has been studied for the period of interest at location 12L.

The results obtained for the sanitary waste parameters are shown in Table 29. The averages for January, February, and December exceeded the State of Illinois standard for ammonia nitrogen. The five-day biochemical oxygen demand and dissolved solid values were below the standards.

TABLE 29

Performance of Sanitary Waste Treatment Plant, 1979

Month	Concentrations (mg/l)		
	B.O.D. ₅	Ammonia Nitrogen	Suspended Solids
January	4.6	9.7	6.3
February	3.3	8.2	6.0
March	2.0	2.9	3.1
April	3.1	2.3	2.0
May	3.5	0.4	6.5
June	2.1	0.6	4.8
July	1.0	0.02	4.9
August	2.6	0.04	4.7
September	1.3	0.3	2.5
October	1.6	1.3	2.0
November	0.8	3.8	6.3
December	5.8	6.7	3.7
State Standard	10	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	15

The results obtained for chemical constituents in the waste treatment plant effluent are shown in Table 30. All of the average concentrations were at or below the State standards. The average value for mercury was 78% of the State standard and 16% of the samples exceeded this value. Efforts are continuing to lower these levels. The level of hexavalent chromium was non-detectable. This is indicative of the removal of hexavalent chromium from some cooling tower systems as well as the shutdown of the CP-5 reactor, which was a major source of this effluent. The levels of trivalent chromium are approaching natural levels.

With the increased sensitivity now available for barium analyses, results truly representative of background levels are seen. Levels for arsenic, beryllium, cadmium, fluoride, iron, lead, manganese, nickel, silver, and zinc are similar to past years' results and are probably a measurement of background

TABLE 30

Chemical Constituents in Effluent From ANL Treatment Plant, 1979

Constituent	No. of Samples	Concentration ($\mu\text{g/l}$)			Percent of Standard (Avg.)	Percent Exceeding State Standard
		Avg.	Min.	Max.		
Arsenic	56	< 10	-	< 10	< 4	0
Barium	56	14 ± 1	6	25	0.7	0
Beryllium	10	< 0.04	-	0.05	-	-
Cadmium	56	1.0 ± 0.1	< 0.4	2.8	0.7	0
Chromium(VI)	56	< 10	< 10	20	< 3	0
Chromium(III)	56	9 ± 4	< 6	22	9	0
Copper	56	43 ± 8	20	231	4.3	0
Fluoride	56	279 ± 27	190	800	1.9	0
Iron	56	235 ± 10	150	330	12	0
Lead	56	3 ± 0.4	< 2	8.8	3	0
Manganese	56	23 ± 5	7	106	2.3	0
Mercury	250	0.39 ± 0.05	< 0.1	3.4	78	16
Nickel	56	8 ± 2	< 3	59	0.8	0
pH	250	-	6.7	7.6	-	0
Selenium	56	< 10	-	< 10	< 1	0
Silver	56	3 ± 0.3	1	7	3	0
Zinc	56	200 ± 35	100	786	20	0

levels. It is interesting to note that levels of zinc have not changed even though zinc compounds have been used to partially replace the chromate inhibitor. The average level of copper is about the same as last year's value.

Results obtained for the cooling tower effluents are shown in Table 31. Except for 8J, average chromium concentrations at all stations were below the detection limits. The cooling towers which provide blowdown for the 8J stream are being switched from chromate treatment to a combination of zinc compounds. Monitoring for zinc as well as chromate has begun at this location. Zinc levels at the combined effluent, 12L, from June through December, have remained very low and do not appear to result from other than natural causes. It is anticipated the 1980 will see the end of chromate contamination at all effluents. Careful surveillance of zinc levels will continue.

b. Sawmill Creek

Samples collected for evaluation of the effect of the sanitary wastes on stream quality were obtained once per week using specially constructed sampling bottles. The sampling bottles were designed to provide temperature measurements as well as to minimize changes in oxygen content during collection.

These samples were collected 15 m (50 feet) upstream of the Argonne outfall [7M (up)] and 60 m (200 feet) downstream of the outfall [7M (down)]. Additionally, samples to be examined for fecal coliform were collected at least five times per month at both 7M locations, but downstream the samples were collected immediately in front of the outfall grating to minimize contamination from Sawmill Creek. Once per month a sample was obtained as the water enters the site (16K), which is downstream of the Marion Brook Treatment Plant.

Except for fecal coliform, the data from these studies are in Table 32. The average level for ammonia nitrogen upstream is 1.5 times the State of Illinois standard and individual samples exceeded the standard 59% of the time. The downstream sample averaged 1.2 times the State standard and exceeded this standard 49% of the time. All of the samples obtained downstream that exceeded the standard were due to upstream contamination. The dissolved oxygen levels obtained during 1979 averaged 108% and 110% saturation for 7M (up) and 7M (down) samples. The fact that values exceeded 100% saturation is

TABLE 31

Cooling Tower Effluents, 1979

Constituent	Location *	No. of Samples	Concentration ($\mu\text{g/l}$)			Percent of Standard (Avg.)	Percent Exceeding State Standard
			Avg.	Min.	Max.		
Chromium(VI)	8J	44	1410 \pm 300	10	4000	470	84
Chromium(VI)	11L	22	< 10	-	< 10	< 3	0
Silver	11L	21	1.3 \pm 0.4	< 0.2	2.8	1.3	0
Chromium(III)	11L	21	3.6 \pm 1.4	< 1	13.5	4.5	0
Chromium(VI)	12F	35	< 10	-	< 10	< 3	0
Chromium(VI)	12L	43	< 10	-	< 10	< 3	0
Zinc	12L	25	25.0 \pm 2.4	< 20	38	2.5	0
Chromium(VI)	14G	34	< 10	-	< 10	< 3	0
Chromium(VI)	14H	30	< 10	-	< 10	< 3	0
Chromium(VI)	14J	34	< 10	-	< 10	< 3	0

* Locations are given in terms of the grid coordinates in Figure 1.

TABLE 32

Sawmill Creek - Effect of Sanitary Waste, 1979

Constituent	Location *	No. of Samples	Concentration (mg/l)			Percent of Standard	Percent Exceeding State Standard
			Avg.	Min.	Max.		
Ammonia	7M (up)	39	2.3 \pm 0.7	0.1	9.4	153	59
Nitrogen	7M (down)	39	1.8 \pm 0.5	0.1	6.6	120	49
Dissolved Oxygen**	7M (up)	45	108 \pm 7	47	155	-	-
	7M (down)	45	110 \pm 5	69	143	-	-
Temperature °C	7M (up)	45	15.8 \pm 1.9	4.7	26.6	-	-
	7M (down)	45	16.7 \pm 1.7	6.2	26.3	-	-
Total	7M (up)	45	1184 \pm 136	446	2085	118	58
Dissolved Solids	7M (down)	45	954 \pm 85	458	1617	95	53

* Location 7M (up) is 15 m (50 ft) upstream from the waste-water outfall. All other samples were collected 60 m (200 ft) downstream from the outfall.

** Percent saturation at measured temperature.

due primarily to photosynthetic activity. The total dissolved solids above the outfall exceeded the State standard frequently, and increased the solids content of the downstream samples. Similar results for these constituents were obtained in 1978.

The fecal coliform standard requires that the monthly geometric mean not exceed 200 organisms/100 ml and that no single sample exceed 400 organisms/100 ml. Samples obtained above the 7M outfall exceeded the 200 organisms/100 ml standard in February and May, and there were eight instances when the 400 organisms/100 ml standard was exceeded. The sample obtained at 16K exceeded the 400 organisms/100 ml standard in June, July, August, and September. Samples obtained at the Argonne outfall never exceeded the 200 organisms/100 ml monthly standard nor the 400 organisms/100 ml standard.

Samples to evaluate the effect of combined sanitary and laboratory waste on the concentrations of chemical constituents in Sawmill Creek were collected by a continuous sampler in the Creek five times per week. These were the same samples taken for radioactivity analyses. The results are summarized in Table 33.

Individual samples for arsenic, cadmium, chromium, cyanide, fluoride, lead, manganese, nickel, and zinc did not exceed the State standards. Previously elevated hexavalent chromium levels have been reduced to non-detectable levels, and during this period, only one sample contained measurable amounts. The source for this sample has been shown to be upstream of Argonne, since a sample obtained at location 16K had a sufficiently high hexavalent chromium content to account for the levels found in the downstream sample. During 1979, then, no samples contained measurable amounts of hexavalent chromium due to Argonne operations. The levels of mercury averaged 44% of the State limit and exceeded this limit 8% of the time. Results for barium represent an accurate description of barium levels in this stream. Interferences of calcium and/or dissolved solids have been better controlled than previously and very high sensitivity resulting from flameless techniques has yielded realistic results for barium. Selenium is reported for the first time and results are low. As in the past, high iron levels reflect high turbidity conditions and are not related to Argonne releases. One silver sample exceeded the State limit by 20%. The levels of copper exceeded State limits 54% of the time. A careful study of both upstream and downstream levels has

TABLE 33

Chemical Constituents in Sawmill Creek, Location 7M,* 1979

Constituent	No. of Samples	Concentration ($\mu\text{g/l}$)			Percent of Standard (Avg.)	Percent Exceeding State Standard
		Avg.	Min.	Max.		
Arsenic	52	< 10	-	< 10	< 1	0
Barium	52	52 \pm 7	16	132	1	0
Beryllium	10	0.094 \pm 0.047	< 0.04	0.270	-	-
Cadmium	52	1.3 \pm 0.2	< 0.4	2.7	2.6	0
Chromium(VI)	52	< 10	< 10	240	< 20	2
Chromium(III)	52	24 \pm 9	6	154	2.4	0
Copper	52	36 \pm 8	6	161	180	54
Cyanide	45	< 20	-	< 20	< 80	0
Fluoride	52	340 \pm 30	200	800	24	0
Iron	52	1310 \pm 510	180	1040	131	31
Lead	52	13.6 \pm 4.6	< 2	86	13.6	0
Manganese	52	159 \pm 40	30	880	14.5	0
Mercury	236	0.22 \pm 0.04	< 0.1	4.3	44	8
Nickel	52	9.9 \pm 2.3	< 3	42.2	1	0
pH	236	-	7.2	8.3	-	0
Selenium	46	< 10	-	< 10	< 10	0
Silver	52	2.1 \pm 0.3	0.4	5.8	42	2
Zinc	52	120 \pm 30	40	570	12	0

* Location 7M is 60 m (200 ft) downstream from the waste-water outfall.

shown that water entering above the sampling location contains about 10 $\mu\text{g}/\text{l}$ of copper as compared to the annual average of 36 $\mu\text{g}/\text{l}$ in the downstream sample. The average levels in the Argonne effluent, 43 $\mu\text{g}/\text{l}$, with a one-to-one dilution with Creek water, would produce levels of about 25 $\mu\text{g}/\text{l}$. The average dilution during 1979 was about 10 volumes of Creek water to one volume of Argonne waste. This should produce a stream level of about 15 $\mu\text{g}/\text{l}$, considerably less than the average of 36 $\mu\text{g}/\text{l}$. An additional piece of evidence for a non-ANL source of copper is seen in the data for September. This month was the driest in history for this area. This is reflected in the upstream water volume for September of a daily average of 19 megaliters/day (5×10^6 gal/day) as compared to the 1978 average of 38 megaliters/day (10×10^6 gal/day). The Creek levels of 20 $\mu\text{g}/\text{l}$ during this period of lowest natural dilution were the second lowest of any month in 1979. All of this indicates that, while Argonne did release an annual average of 43 $\mu\text{g}/\text{l}$, copper levels obtained in the stream sample are composed of an upstream background and adventitious copper as well as Argonne releases. A study has begun to evaluate this adventitious component which is felt to be entrainment of stream bottom materials in the sampler used rather than an indication of effluent contamination.

c. Des Plaines River

The effect of Sawmill Creek on the Des Plaines River was evaluated by collecting samples at Willow Springs (upstream of Argonne) and at Lemont (downstream of Argonne). These samples were analyzed for total mercury, hexavalent chromium, total iron, and total zinc. The results are in Table 34. Two of the samples had very high levels of suspended material, as evidenced by iron levels of 1.9 and 3.2 mg/l. In no case was there any indication that hexavalent chromium, mercury, or zinc levels were affected by the Argonne effluent.

2. Environmental Air Studies

Studies of background levels of total suspended particulates were continued during 1979 at four locations situated to study effluents from the Argonne power plant. These samples were also analyzed for iron, copper, zinc, lead, bromine, and strontium using x-ray fluorescence spectrometry.

Samples for particulate matter determination were collected on glass

TABLE 34

Chemical Constituents in the Des Plaines River, 1979

Constituent	Location *	No. of Samples	Concentration (mg/l or µg/l)		
			Avg.	Min.	Max.
Chromium(VI)	A	12	< 0.01	-	< 0.01
	B	23	< 0.01	< 0.01	0.04
Iron	A	12	1.0 ± 0.4	0.3	1.9
	B	23	1.0 ± 0.3	< 0.1	3.2
Mercury **	A	12	< 0.1	-	< 0.1
	B	23	< 0.1	-	< 0.1
Zinc	A	12	0.04 ± 0.01	0.02	0.07
	B	23	0.04 ± 0.01	0.02	0.12

* Location A, near Route 45, is upstream and Location B, near Lemont, is downstream from the mouth of Sawmill Creek. See Figure 2.

** Concentrations in µg/l.

fiber filters (20 cm x 25 cm). High volume samplers (General Metals Corp.) were used and samples were collected for seven-day periods. The air flow rate varied from 1.1 to 1.9 m³/min., depending on the sampler design, and was verified using a Roots meter as a primary standard. The papers were equilibrated before and after use in a dry box (relative humidity less than 35%) for a minimum of 24 hours.

Sample location 14N is in the Waterfall Glen Forest Preserve in an area with very low traffic volume. The sampler is located approximately 4 m (13 ft) above the ground and is frequently in a downwind direction of the Argonne power plant.

Sample location 12F is west of the power plant and is approximately 3 m (10 ft) above ground. Sample location 8F is southwest of the power plant, at the Argonne meteorology station, and is 1.3 m (4 ft) above the ground. This sampler also monitors effluent from power plants located southwest of the Laboratory. Sample location 12M is southeast of the power plant and located about 6 m (20 ft) above ground. This is the nearest sampler to the plant. Beginning October 24, 1979, a 24-hour sample was collected every six days at location 12M and handled in the same fashion.

Trace element analyses were performed on the samples collected for total suspended particulates by removing a one centimeter circle and counting it for one hour using a cadmium-109 source to excite the x-rays. Studies have shown that distribution of these six elements across the filter is sufficiently homogenous so that counting this segment gives results representative of the entire filter. Due to a variety of problems, results are available for only the first quarter of 1979.

The results are shown in Table 35. All values are listed as geometric means since the Federal Standard is in these terms. Figure 7 is a log-normal plot of the TSP data from location 12F. The fit of the data to the least-squares line indicates this distribution is a good description of the data.

The lack of trace metal data precludes an extensive correlative study. This discussion will be included in the 1980 annual report.

The levels of total suspended particulates and trace elements reported herein are normal constituents of ambient air and represent background levels which will be useful for evaluating changes due to planned future coal-burning at Argonne.

TABLE 35

Environmental Air Data
(Concentrations in $\mu\text{g}/\text{m}^3$) *

Constituent	Location			
	14N	12F	8F	12M
Total Suspended Particulates	43	55	45	45
Iron	0.87	1.0	0.91	0.95
Copper	0.09	0.08	0.13	0.21
Lead	0.46	0.50	0.46	0.44
Zinc	0.19	0.22	0.19	0.18
Bromine	0.10	0.11	0.10	0.09
Strontium	0.002	< 0.001	0.003	0.002
12M - 24-hour particulate - $36 \mu\text{g}/\text{m}^3$.				

* Average geometric mean.

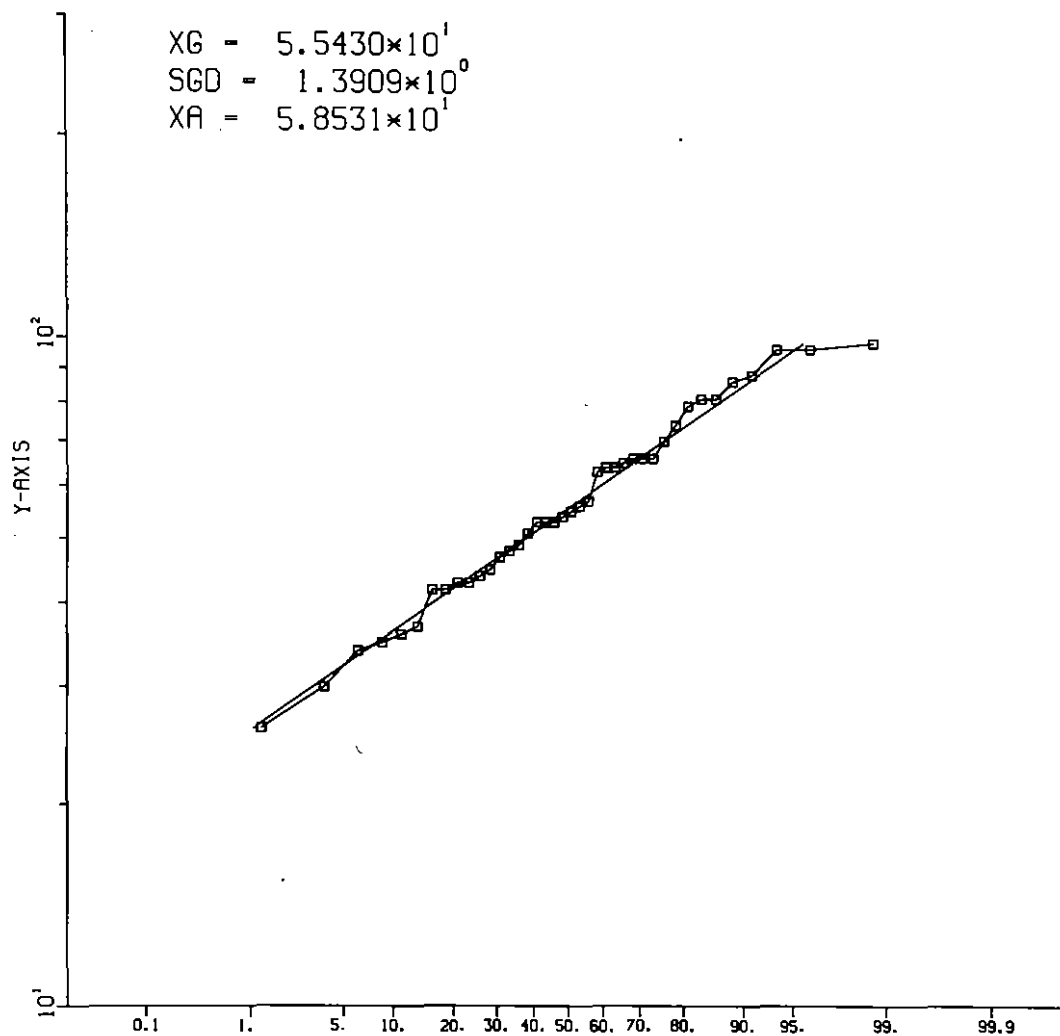


Fig. 7. Log-normal Distribution Plot of Lead Content of Air at ANL 1979. Geometric mean - $0.39 \mu\text{g}/\text{m}^3$; geometric standard deviation - $1.54 \mu\text{g}/\text{m}^3$; arithmetic mean - $0.43 \mu\text{g}/\text{m}^3$. The straight line is a least-squares fit to the data points.

IV. APPENDIX

A. References

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B. Environmental Quality Standards, Detection Limits, Analytical Errors, and Quality Assurance

1. Standards

All of the standards and detection limits for chemical constituents, and some of those for radionuclides and external radiation, are given in the main body of the report. In addition, in Table 36 are collected air and water environmental quality standards and detection limits (minimum detectable amounts) for all radionuclides, and for those materials, for which measurements were made. These standards are the Concentration Guides given in DOE Manual Chapter 0524, ⁽⁴⁾ and are used in this report to assess the hazard of a measured concentration of a radioactive nuclide. Chapter 0524 distinguishes two CGs, one for occupational exposure in controlled areas and one for uncontrolled area, beyond the site boundary where individuals can be exposed nonoccupationally, for 168 hours per week. The CGs in the table are for uncontrolled areas. For water, the standard selected was for the soluble form of the radionuclide; for air, the standard for the insoluble form was selected (except for iodine-131, for which the soluble form was chosen as a more conservative standard).

TABLE 36

Concentration Guides and Detection Limits
($\mu\text{Ci/ml}$)

Nuclide or Activity	Concentration Guide		Detection Limit	
	Water	Air	Water	Air
Americium-241	4×10^{-6}	4×10^{-12}	1×10^{-12}	5×10^{-19}
Antimony-125	-	9×10^{-10}	-	5×10^{-16}
Argon-41	-	4×10^{-8}	-	1.5×10^{-8}
Barium-140	3×10^{-5}	1×10^{-9}	2×10^{-9}	5×10^{-16}
Beryllium-7	-	4×10^{-8}	-	5×10^{-15}
Californium-249	4×10^{-6}	-	1×10^{-12}	-
Californium-252	7×10^{-6}	-	1×10^{-12}	-
Cerium-141	-	5×10^{-9}	-	5×10^{-16}
Cerium-144	-	2×10^{-10}	-	1×10^{-15}
Cesium-137	2×10^{-5}	5×10^{-10}	-	5×10^{-16}
Cobalt-60	-	3×10^{-10}	-	1×10^{-16}
Curium-242	2×10^{-5}	-	1×10^{-12}	-
Curium-244	7×10^{-6}	-	1×10^{-12}	-
Hydrogen-3	3×10^{-3}	2×10^{-7}	1×10^{-7}	1×10^{-13}
Iodine-131	3×10^{-7}	1×10^{-10}	3×10^{-9}	5×10^{-15}
Manganese-54	-	1×10^{-9}	-	5×10^{-16}
Neptunium-237	3×10^{-6}	-	1×10^{-12}	-
Plutonium-238	5×10^{-6}	1×10^{-12}	1×10^{-12}	1×10^{-19}
Plutonium-239	5×10^{-6}	1×10^{-12}	5×10^{-13}	1×10^{-19}
Ruthenium-103	-	3×10^{-9}	-	5×10^{-16}
Ruthenium-106	-	2×10^{-10}	-	1×10^{-15}
Strontium-89	3×10^{-6}	1×10^{-9}	2×10^{-9}	1×10^{-16}
Strontium-90	3×10^{-7}	2×10^{-10}	2.5×10^{-10}	1×10^{-17}
Thorium-228	-	2×10^{-13}	-	1×10^{-18}
Thorium-230	-	3×10^{-13}	-	1×10^{-18}
Thorium-232	-	1×10^{-12}	-	1×10^{-18}
Uranium-234	-	4×10^{-12}	-	1×10^{-19}
Uranium-235	-	4×10^{-12}	-	1×10^{-19}
Uranium-238	-	5×10^{-12}	-	1×10^{-19}
Uranium - natural*	4×10^{-5}	4×10^{-12}	2×10^{-10}	2×10^{-17}
Zirconium-95	-	1×10^{-9}	-	5×10^{-16}
Alpha**	3×10^{-6}	1×10^{-10}	2×10^{-10}	2×10^{-16}
Beta**	to 1×10^{-7}	to 1×10^{-13}	1×10^{-9}	5×10^{-16}

* Concentration Guides converted from the "special curie" used in DOE M 0524 to the standard curie.

** The Concentration Guides for unknown mixtures depend, within the range given, on whether certain radionuclides are known to be present in concentrations less than 0.1 of their CGs, and the sum of the fraction of the CGs for all such nuclides is less than 0.25. For most total alpha and beta results given in this report, the largest uncontrolled CG is applicable.

2. Detection Limits

The detection limits were chosen so that the error at the 95% confidence level is equal to the detection limit. The relative error in a result decreases with increasing concentration. At a concentration equal to twice the detection limit, the error is about 50% of the measured value, and at 10 times the detection limit, the error is 10%.

3. Quality Assurance Program

a. Radiochemical Analysis and Radioactivity Measurements

All nuclear instrumentation is calibrated with standard sources obtained from the U. S. National Bureau of Standards (NBS), if possible. If NBS standards were not available for particular nuclides, standards from the Amersham-Searle Co. were used. The equipment is usually checked on a daily basis with secondary counting standards to insure proper operation. Samples are periodically analyzed in duplicate or with the addition of known amounts of a radionuclide to check precision and accuracy. In addition, standard and intercomparison samples distributed by the DOE Environmental Measurements Laboratory (EML), the Quality Assurance Branch of the U. S. Environmental Protection Agency (EPA-QA) at Las Vegas, and the International Atomic Energy Agency (IAEA) are analyzed regularly. The EPA-QA intercomparison program consists of analyzing a variety of samples, at intervals selected by the participant, to which known amounts of various radionuclides have been added by the EPA laboratory. A summary of all the EPA-QA samples analyzed in this laboratory in 1979 is shown in Table 37. The DOE Environmental Measurements Laboratory Quality Assurance Program (DOE-EML-QAP) is a quarterly distribution of four or five different sample matrices containing various combinations of radionuclides.⁽¹⁶⁾ Results of our participation in this program during 1979 are given in Table 38. In the table, the comparison is made between the EML value, which is the result of replicate determinations by that Laboratory, and the value obtained in our laboratory. More than 95% of all the intercomparison samples received were analyzed for the radionuclides for which results were requested. To assist in judging the quality of the results, typical errors for our analyses are 2-50%, the error in the EML results is 1-30% (depending on the nuclide and the amount present), and the error in the added amount in the EPA-QA samples is 2-5% (our estimate).

TABLE 37

Summary of EPA-QA Samples, 1979

Type of Sample	Analysis	Number Analyzed	Avg. Difference From Added
Air Filter	Total Alpha	3	31%
	Total Beta	3	9%
	Strontium-90	2	7%
	Cesium-137	3	11%
Water	Total Alpha	6	36%
	Total Beta	6	9%
	Hydrogen-3	3	8%
	Chromium-51	1	4%
	Cobalt-60	3	6%
	Zinc-65	1	5%
	Strontium-89	2	1%
	Strontium-90	2	15%
	Iodine-131	2	6%
	Cesium-134	3	8%
	Cesium-137	2	6%
	Radium-226	2	51%
	Radium-228	1	9%
	Total Uranium	1	13%
	Plutonium-239	2	9%
Milk	Potassium-40	2	3%
	Strontium-90	1	11%
	Iodine-131	2	4%
	Cesium-137	2	7%

TABLE 38

Summary of DOE-EML-QAP Samples, 1979

Nuclide	Average Difference From EML Value				
	Air Filters	Water	Soil	Tissue	Vegetation
Hydrogen-3	-	6% (3)	-	-	-
Beryllium-7	18% (2)	-	-	-	-
Sodium-22	5% (1)	2% (2)	-	-	-
Potassium-40	-	-	15% (4)	14% (3)	8% (3)
Manganese-54	2% (1)	4% (1)	-	-	-
Cobalt-57	2% (1)	9% (1)	-	-	-
Cobalt-58	8% (1)	-	-	-	-
Cobalt-60	4% (1)	3% (3)	3% (1)	-	-
Zinc-65	-	13% (1)	-	-	-
Strontium-89	4% (2)	10% (2)	-	-	-
Strontium-90	33% (3)	6% (2)	5% (3)	3% (1)	27% (2)
Zirconium-95	4% (2)	-	-	-	-
Ruthenium-106	28% (2)	-	-	-	-
Antimony-125	5% (2)	-	-	-	-
Cesium-134	14% (2)	11% (2)	-	-	-
Cesium-137	9% (3)	4% (4)	7% (3)	14% (2)	12% (3)
Cerium-144	2% (1)	3% (1)	-	-	-
Radium-226	-	-	9% (4)	10% (1)	20% (2)
Thorium-228	-	-	6% (3)	-	-
Uranium-234	25% (2)	10% (2)	41% (3)	-	-
Uranium-238	13% (2)	14% (2)	37% (3)	-	-
Uranium-total	13% (1)	8% (2)	-	-	-
Plutonium-238	16% (1)	18% (3)	16% (2)	100% (1)	-
Plutonium-239	15% (4)	26% (3)	23% (4)	26% (2)	7% (2)
Americium-241	11% (4)	15% (3)	51% (1)	-	-

The figure in parentheses is the number of samples.

b. Penetrating Radiation

Our laboratory participated in the Fourth International Comparison of Environmental Dosimeters conducted by the DOE Environmental Measurements Laboratory and the School of Public Health, University of Texas (Houston). Participants supplied three sets of each type of dosimeter they wished to evaluate. One set was given a three-month field exposure to natural radiation in Houston; one set was exposed in the laboratory to a known (but unrevealed) quantity of radiation approximately equivalent to a three-month natural field exposure; and one set was exposed to a known quantity of radiation in a higher range. The latter two exposures were to a radium-226 source. After exposure, the dosimeters were returned to the participant for measurement. The natural radiation field exposure was measured by an EML high-pressure ion chamber for comparison. The results, in mR, were:

	<u>Field Exposure</u>	<u>Laboratory Exposure</u>	
		<u>Low</u>	<u>High</u>
School of Public Health	14.1 ± 0.7	12.2 ± 1.2	45.8 ± 4.6
ANL Value			
calcium fluoride	12.9 ± 2.4	11.7 ± 2.0	44.1 ± 5.9
lithium fluoride	14.6 ± 2.7	10.7 ± 2.7	43.7 ± 2.9

The uncertainties listed are the standard deviations as estimated by the School of Public Health for their values and by us for our results. Our standard deviations are relatively high because substantial corrections were required for irradiation of the dosimeters during storage and during air shipment. All results agree with the standard or accepted values within the measurement error. The most important comparison is for the field exposures. Here the differences between the ion-chamber result and our results were $1.2 \text{ mR} \pm 2.5 \text{ mR}$ (8.5% \pm 17.7%) for calcium fluoride and $0.5 \text{ mR} \pm 2.8 \text{ mR}$ (3.6% \pm 20%) for lithium fluoride.

c. Chemical Analysis

With each set of atomic absorption analyses, standard amounts of trace metals were analyzed in blank solutions at concentrations corresponding to 50 and 100% of the current State standards. Recoveries were determined by comparing these results to results obtained by analyzing stream and effluent samples to which identical concentrations were added. Average recoveries ranged from about 85-100%, with a standard deviation of about 10%. Recovery

studies were also performed for ammonia nitrogen, fluoride, and hexavalent chromium analyses, and similar results were obtained.

The laboratory also participated in the intercomparison program for the analysis of trace elements in water sponsored by the EPA (Cincinnati Laboratory). The results for the three concentration levels distributed are shown in Table 39. In some cases the concentrations added by the EPA were below the normal limits of detection by direct analysis, e.g., selenium and zinc. In other cases, e.g., beryllium and cadmium, levels were considerably higher than environmental levels normally encountered. The results obtained for all of the elements are in agreement within expected ranges. A flameless atomic absorption technique was used for these determinations, except for arsenic, selenium, and zinc. Conventional flame atomic absorption was used for zinc, and arsenic and selenium were determined using x-ray fluorescence.

d. Sampling, Sample Storage, etc.

Many factors enter into an overall quality assurance program other than the analytical quality control discussed above. Representative sampling is of prime importance. The continuous water sampler in Sawmill Creek provides a representative sample for a critical sampling location since the rate of discharge of waste water varies appreciably during each 24-hour period.

The accuracy of the flowmeters in the air sampling equipment is verified periodically with a calibrated rotameter, as well as by comparison to a Roots meter primary standard.

Samples are pre-treated in a manner designed to maintain the integrity of the constituent sought. For example, samples for trace radionuclide analysis are acidified immediately after collection to prevent hydrolytic loss of metal ions, but aliquots for radioiodine analyses are withdrawn first, since trace iodine is unstable in acid solution.

TABLE 39

Results of EPA Quality Assurance Samples
(Concentrations in $\mu\text{g/l}$)

Metal	Lower Limit of Detection	<u>Found</u> Added #1	<u>Found</u> Added #2	<u>Found</u> Added #3	Avg. <u>Found</u> Added
Arsenic	10	13/24	150/182	53/61	0.73
Beryllium	0.02	24/24	205/261	170/183	0.91
Cadmium	0.40	5/6.5	49/59	22/27	0.80
Chromium	2	3/4.4	315/304	70/65	0.93
Copper	0.8	8.5/8.7	372/374	34/37	0.96
Iron	5	16/16	860/796	84/78	1.05
Lead	2	27/30	404/383	104/113	0.96
Manganese	1	4/7.9	446/478	46/47	0.81
Mercury	0.1	0.4/0.4	7.1/7.6	2.8/4.4	0.86
Nickel	3	6.6/8.7	164/165	88/96	0.99
Selenium	10	8.8/8.7	53/48	18/16	1.08
Zinc	10	< 10/6	471/478	29/26	1.07

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